1971
Quarterly
Technical
Progress
Review
Volume 8-4

Isotopes & Radiation Technology

Prepared for
Division of
Technical
Information —
Division of
Isotopes
Development
U.S. Atomic Energy Commission
by Isotopes
Information
Center
Oak Ridge
National Laboratory

See page 373

XUM

TECHNICAL PROGRESS REVIEWS

The United States Atomic Energy Commission publishes the Technical Progress Reviews to meet the needs of industry and government for concise summaries of current nuclear developments. Each journal digests and evaluates the latest findings in a specific area of nuclear technology and science. *Nuclear Safety* is a bimonthly journal; the other two are quarterly journals.

Isotopes and Radiation Technology
P. S. Baker, A. F. Rupp, and associates
Isotopes Information Center, Oak Ridge National Laboratory

Nuclear Safety
Wm. B. Cottrell, W. H. Jordan, J. P. Blakely, and associates
Nuclear Safety Information Center, Oak Ridge National Laboratory

Reactor Technology
Prepared in cooperation with the American Nuclear Society

All are available from the U. S. Government Printing Office. See the back cover for ordering instructions.

NOTICE

This journal was prepared under the sponsorship of the U. S. Atomic Energy Commission. Neither the United States nor the U. S. Atomic Energy Commission, or any of their employees, contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights.

Availability of Reports Cited in This Review

United States Atomic Energy Commission (USAEC) reports are available at certain libraries that maintain collections of these reports. The libraries are listed on the inside front cover of each issue of *Nuclear Science Abstracts*. USAEC reports are also sold by the following governmental and international organizations: (1) National Technical Information Service (NTIS), U. S. Department of Commerce, Springfield, Va. 22151; (2) International Atomic Energy Agency (IAEA), Vienna, Austria; and (3) National Lending Library, Boston Spa, England.

Other U. S. Government agency reports identified in this journal are generally available from NTIS.

Reports from other countries are generally available at the same U. S. libraries as maintain collections of USAEC reports, and from IAEA and the originating country. **United Kingdom Atomic Energy Authority (UKAEA)** reports are sold by Her Majesty's Stationery Office, London. **Atomic Energy of Canada Limited (AECL)** reports are sold by the Scientific Document Distribution Office, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada. UKAEA and AECL reports issued after March 1, 1967, are sold by NTIS to purchasers in the United States and its territories. IAEA publications are sold in the United States by UNIPUB, P. O. Box 433, New York, N. Y. 10016.

Private-organization reports should be requested from the originator.

The views expressed in this publication do not necessarily represent those of the United States Atomic Energy Commission, its divisions or offices, or of any Commission advisory committee or contractor.

Isotopes and Radiation Technology

Vol. 8, No. 4 Summer 1971

Contents

Anniversary

373 **Twenty-five Years of Radioisotope Distribution and Utilization**
Glenn T. Seaborg

I Isotope Technology Development

377 **Nuclear Techniques and the Fight Against Pollution**
Summarized by Martha Gerrard

379 Supplementary Bibliography

379 **Activation-Analysis and Sulfur-Isotope-Ratio Techniques for Oil-Slick Identification** *Summarized by Martha Gerrard and F. E. McKinney*

379 Experimental Procedure
380 Results
380 Usefulness of Methods

381 **The $^{12}C/^{13}C$ Ratio as an Indicator of Air Pollution** *Patrick R. Atkins*

382 Theory
383 Procedures
383 Results

385 **Comparison of Radioisotopes and Fluorescent Dyes for Estuary Pollution-Control Studies** *Summarized by Helen P. Raaen*

386 Discussion
387 Summary

389 **Tritiated-Water-Vapor Method for Studying Lake Evaporation**
P. J. Barry and W. F. Merritt

389 Principles of Method
390 Experimental Procedure

392 **Radioisotopic Sand-Tracer Study** *Summarized by Martha Gerrard*

392 Sand Labeling
392 Injection Device
393 Field Tests
394 Comparison of RIST with Other Tracing Methods

395 **Radioisotopic Gages in Pollution Control** *Martha Gerrard*

395 Eggshell-Strength Gage
396 Suspended-Sediment Gage
396 Other Gages

397 **Measurement of Sulfur Dioxide in Stack Gases: Design, Fabrication, and Test of Demonstration Instrument**
Summarized by R. H. Lafferty, Jr., P. S. Baker, and Martha Gerrard

398 System Description
398 Instrument Construction and Operation
400 Testing
401 Conclusions
402 Appendix: Nonnuclear Methods for Sulfur Dioxide Determination

XUM

403 **Kryptonate-Based Instrument for Detecting Automobile-Exhaust Pollutants**
Summarized by Martha Gerrard and R. H. Lafferty, Jr.
404 Detection System
404 Tests
406 System Advantages
406 Appendix: Nonnuclear Methods for Air-Pollution Monitoring
408 **Production of Synthetic Fallout at Camp Parks** *William B. Lane*
409 Synthetic-Fallout-Production Process
410 Synthetic-Fallout Facilities
412 **Detection System for Tracing Radionuclide-Labeled Sediment in the Marine Environment** *F. N. Case, E. H. Acree, and H. R. Brashear*
412 System Description
413 System Performance

II **Process Radiation Development**

415 **Potential Role of Radiation in Waste-Water Treatment** *D. S. Ballantine*
416 Radiation-Initiated Reactions
419 Sedimentation of Solids
419 Destruction of Bacteria
420 **High-Energy Radiation Synthesis of Ozone for Water Treatment**
Meyer Steinberg and Morris Beller
421 Basic Energy Requirements
422 History
422 Ozone Synthesis Tests
423 Radiation Process for Ozone Production
424 Economics of Radiation Process for Ozone Production
429 **Sewage and Waste-Water Processing with Isotopic Radiation: Survey of the Literature** *Martha Gerrard*
429 Sedimentation of Suspensions
430 Disinfection
431 Destruction of Organic and Inorganic Compounds
432 Economics
435 **Conceptual Design of an Irradiation Test Facility for Waste Water and Sewage Sludge** *Summarized by Martha Gerrard*
436 Design
436 Radiation Source
438 Cost
439 **Biological–Gamma-Radiation System for Sewage Processing** *Leland A. Mann*
439 Pilot Plant
439 Palmdale Plant
441 Costs
443 Radiation Safety
444 **Radiation Treatment of Industrial Waste Waters: An Economic Analysis**
A. I. Mytelka
445 Economic Analysis
448 Discussion
449 Conclusions
449 **Gamma Irradiation as a Pretreatment to Chemical Precipitation in the Purification of Domestic Sewage** *Lorne A. Campbell*
449 Procedure
450 Results

451 **Gamma Irradiation of Waters and Waste Waters for Disinfection Purposes**
A. H. Vajdic
451 Disinfection
451 Taste and Odor Removal
452 **Gamma Irradiation of Raw Sewage for Sedimentation Purposes**
R. H. Andrews and M. B. Fielding
452 Procedure
452 Results
453 **Destruction of Organic Substances in Waste Water by Ionizing Radiation**
D. M. J. Compton
453 Theoretical Considerations
455 Results
456 Description of Radiation Source

III Miscellaneous

459 **Recent Reports on Pesticides**
460 **Pollution Problems**
461 **Availability of Isotopes and Services**
463 **General**
466 **Abstracts of Recent Literature**

Index

476 **Volume 8**

Superintendent of Documents, U. S. Government Printing Office, Washington, D. C. 20402, $2.50 per year.

XUM

Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

Insofar as feasible all references are checked by the editors, using the library facilities of Oak Ridge National Laboratory, Oak Ridge Associated Universities, and the University of Tennessee. Occasionally, however, the cited material is not available; such cases are identified by an asterisk (*).

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Heat and Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

P. S. Baker, R. H. Lafferty, Jr., Martha Gerrard, R. E. Greene, F. E. McKinney, and Helen P. Raaen, *Editors*
A. F. Rupp, *Adviser*

OAK RIDGE NATIONAL LABORATORY

Issued quarterly by the U. S. Atomic Energy Commission. Use of funds for printing this publication approved by the Director of the Bureau of the Budget on December 16, 1968.

Twenty-five Years of Radioisotope Distribution and Utilization

By Glenn T. Seaborg*

The 25th anniversary of the first radioisotope shipment is an appropriate time to reflect on what has been accomplished in the production and application of these materials which now have important uses in the fields of environment and space as well as medicine, industry, and agriculture. This first peacetime product of atomic energy had its formal beginning on Aug. 2, 1946, when a group of scientists and newsmen gathered at Clinton Laboratories in Oak Ridge, Tenn., to witness the first shipment of reactor-produced radioisotopes.

It was by chance that Barnard Free Skin and Cancer Hospital of St. Louis, Mo., cleared the elaborate procurement procedures to receive the first shipment. The radioisotope received by the hospital was for research and study of the processes by which cancer is produced. Many others were waiting to receive shipments for use in such widely divergent research projects as vulcanization and polymerization of rubber, utilization of foods by animals, and absorption of nutrients from the soil by plants.

This sudden demand for radioisotopes was generated in large measure by earlier work with radium and the small quantities of cyclotron-produced radioisotopes that could be made. The uranium piles, which had so successfully fulfilled their purpose during World War II, made it possible to have radioisotopes in abundance—and at comparatively small cost. Thus the first radioisotopes catalog published in *Science*, June 14, 1946, was eagerly received by scientists and engineers returning to their normal areas of endeavor.

The first shipment was 1 mCi of ^{14}C as barium carbonate, and it weighed about 3 mg. Despite its small physical size, this represented nearly 1000 times as much of the isotope as had previously been made available for research in any single order of cyclotron-produced ^{14}C. Today we have a capability for routinely producing many more kinds of radioisotopes than was possible in 1946—and in quantities millions of times greater than the first shipment. The efforts of industry, the medical profession, scientists, and government laboratories to develop new nuclear techniques have indeed been substantial. Applications of radioisotopes have increased until today they are almost commonplace in hospitals, industrial plants, and research establishments around the world.

ISOTOPE PRODUCTION

Much effort has been spent in a continuing search for new and better isotopes. Particular attention has been paid to the needs in medical research, diagnosis, and therapy. Clinical-grade radioisotopes must be as free as possible from contaminants and available in different forms to meet many specialized needs.

Much attention also has been given to the engineering development of industrial radiation sources. In some cases this has involved radiation intensities of millions of roentgens per hour at high thermal temperatures. It is imperative that such sources be leakproof over long periods of time. Also, one must anticipate accidents and assure that radiation workers and the public are protected.

To cope with special manufacturing problems, an Isotopes Development Center was established at Oak Ridge in 1960. In addition, a High Intensity Radiation Development Laboratory was established at Brookhaven National Laboratory in 1962 to give particular attention to the design and utilization of irradiators.

*Chairman, U. S. Atomic Energy Commission.

XUM

During all these product-development operations, AEC has followed a policy of "getting out of the business" as soon as private industry develops a capability to take over. For example, during the past decade AEC has withdrawn from the production and supply of 65 different radioisotopes, in view of private capabilities to meet market demand.

ENVIRONMENTAL APPLICATIONS

When radioisotopes became available in 1946, there was an abundance of everyday problems to challenge the ingenuity of scientists and engineers. These versatile materials were used to seek all sorts of new scientific knowledge. However, problems of environmental control were not among the major issues at that time. These are of more recent vintage and are now perhaps the most urgent issue to arise since radioisotopes became available. The rapidly deteriorating quality of air and water resources is a matter of worldwide concern today. In an effort to better understand and control pollution, radioisotopes and nuclear techniques are being called upon for an important role. The diverse ways in which they can be used is the subject of this special environmental edition of *Isotopes and Radiation Technology*.

MEDICAL APPLICATIONS

Medicine and biology have received the greatest benefits from the ready availability of radioisotopes. As tracer atoms they are uniquely suited to solution of complicated research problems in these areas. Body processes, in sickness and in health, are fundamentally chemical but are carried out on such a dynamic, complex, and microscopic scale that they frequently defy ordinary chemical procedures in their study.

Medical diagnostic techniques also have been greatly extended through the use of radioisotopes. In medical therapy they have come into wide utilization for teletherapy, surface applicators, and in situ irradiation of such selected body tissues as the thyroid gland. The in situ mode of treatment is unique in that body processes will concentrate selected forms of radioactive atoms in certain tissues for localized irradiation.

The growing importance of isotopes in medicine is illustrated by the fact that some 5000 U. S. hospitals are now using nuclear medicine. About 2500 physicians are licensed to use radioisotopes in private practice. The annual market for nuclear medicine and associated equipment totals close to $70 million, exclusive of radiochemicals and sealed sources. To date some 100 different radioisotopes have been used in medical research. Thirty different radioisotopes are available in quantity for medical purposes. In the United States alone, radioisotopes are used in approximately eight million individual diagnostic and therapeutic treatments a year.

The list of various radioisotopes and their medical uses is long. More than one million ^{131}I "atomic cocktails" are served in the United States each year for the thyroid-function test. Versatile ^{99m}Tc is used in the diagnosis of thyroid, liver, brain, and kidney diseases.

Work is going ahead on development of an artificial heart with ^{238}Pu selected as the power source. Another exciting prospect is the powerful and versatile new radioisotope ^{252}Cf that has the useful property of emitting neutrons as part of its decay process. Neutrons may be more efficient in destroying oxygen-deficient cancer cells than are X rays and gamma rays. The use of ^{252}Cf in cancer therapy could prove to be a major advancement in the treatment of this disease.

AGRICULTURAL APPLICATIONS

The farmer, faced with the task of feeding a growing population, has used radioisotopes to create better and more productive strains of food crops, to improve crop cultivation and fertilization, and to analyze and thereby improve the care and feeding of livestock.

Nearly 100 new plant strains, including many basic food crops, have been developed in the United States and abroad through mutation by radiation. These include bread wheat resistant to rust diseases, barley with increased winter hardiness, and a peanut strain featuring damage-resistant hulls, good taste, and higher yield.

Through isotopes, plant scientists are now able to better understand the complex photosynthetic process. Progress has been made toward the design of an effective synthetic light-energy conversion process and the genetic engineering of new crop plants of high yield which are closely adapted to particular growth conditions.

By using fertilizer nutrients tagged with isotopes, the scientist can now determine the availability of elements in soils, the best methods and timing of fertilizer applications, the residual value of fertilizers, and the interaction between different fertilizer nutrients.

An important breakthrough in efforts to cut down use of insecticides—and thereby reduce environmental pollution—came with the radiation-sterilized insect release method. In this "sterile male technique," the pest is mass produced in the laboratory, exposed to just enough radiation to render it sterile, then released to infested areas. The sterile pest competes effectively with its wild counterpart and mates but produces no offspring, and the pest population dwindles. In the Southeast, applications of the technique have virtually eliminated the screwworm that infests livestock. A similar campaign is under way against the screwworm in the Southwest and elsewhere against other insects that threaten livestock and crops in the United States and abroad.

INDUSTRIAL APPLICATIONS

Radioisotopes have come into wide industrial utilization to measure and control manufacturing processes; to inspect materials, parts, and finished products; to determine the cause of processing difficulties; and to facilitate research. The techniques are generally less expensive and more effective than the methods and devices they replace. In some cases they have helped industry do things that could not be done by any other means.

Radioisotopes have provided industry with savings or increased profits by decreasing raw-material consumption, reducing finished-product scrap, increasing productivity, and lowering labor costs. They have also speeded up numerous research investigations.

The extent and value of radioisotope applications to industry cannot be accurately assessed. A product that has been materially improved may not only return a higher profit to the maker but may also contribute lower costs and higher value to those who use or resell it. The fact that there are 5300 industrial radioisotope licensees in the United States is indicative of the versatility and value of these materials in industry during the past quarter of a century.

PROCESS RADIATION

The field of process radiation has received much attention, particularly during the past decade. This involves the use of radiation for the initiation of industrially significant chemical reactions or for the processing of commercial materials. Perhaps the most notable progress has been made with wood–plastic composites. Work in this area was started in the early 1960s at West Virginia University. The process involves impregnation of wood products with a liquid monomer and irradiation by gamma rays, during which the monomer hardens as the result of polymerization. The result is a new material with the beauty of wood and the hard, durable properties of plastic. In 1964 a new industrial company was formed for the purpose of commercializing the new wood–plastic material in the form of parquet flooring. Since that date three other companies have entered into commercial production of wood–plastic materials.

FOOD PRESERVATION

Nuclear radiation in large enough doses can destroy bacteria and enzymes in a material without raising its temperature significantly. The availability of large radiation sources has opened interesting new possibilities for food preservation, and much attention has been given to the subject during the past 25 years. This interest was spurred by the fact that many foodstuffs lose their freshness and flavor when subjected to conventional processing such as freezing, drying, and canning.

Radiation pasteurization can control spoilage caused by bacteria, yeasts, and molds in such foods as fresh strawberries, fish, chicken, and citrus fruits and thereby extend their marketable life. It can control insect and parasite infestations in wheat and wheat products and dried fruits, replacing chemical pesticides and their potential residue danger. Radiation energy can slow ripening or aging in fruits and vegetables. Delayed maturation can be achieved for bananas, mushrooms, and mangoes, extending their prime marketing period. Irradiation can also result in the inhibition of sprouting in stored potatoes and onions.

Research has shown that meats exposed to high, or sterilization doses of gamma rays remain edible without refrigeration for up to a year or more. The sterilization technology, which destroys all microorganisms, is aimed at processing perishable foods for long-term storage without refrigeration. Success has been demonstrated in the processing of beef, pork, chicken, smoked ham, frankfurters, codfish cakes, shrimp, ground beef, pork sausages, and canned beef.

Irradiation techniques and effects on individual foods can be complicated, and the Food and Drug Administration (FDA) is proceeding with great caution in granting approvals for commercialization. Wheat and wheat flour and potatoes currently hold FDA approval in the United States, and several items have been approved abroad.

XUM

RADIOISOTOPE POWER

In the early 1960s, serious efforts were begun to develop the use of radioisotopes as small but unfailing sources of power. This led to the development and demonstration of the world's first radioisotope-powered automatic weather station in August 1961 which was placed in service on Axel Heiberg Island in the Canadian Northwest territory.

During man's first journey to the moon, two ^{238}Pu-fueled heaters were placed on the lunar surface on July 21, 1969, by the Apollo 11 astronauts for protection of seismic instrumentation against the cold lunar night.

On Nov. 19, 1969, a radioisotope generator was placed on the moon by the Apollo 12 astronauts to provide electric power for the instrumentation left there. Again, on Feb. 5, 1971, the Apollo 14 astronauts placed another such generator on the moon to power electrical equipment.

These examples serve only to illustrate the unique role radioisotopes have played in meeting very specialized power needs.

CONCLUSION

Radioisotopes have provided the means for some of the major accomplishments of the nuclear age. As technology advances, new doors will be opened for other exciting applications of these versatile materials. They truly have earned the title of "silent servants of mankind."

Section

I

Isotopes and Radiation Technology

Isotope Technology Development

Nuclear Techniques and the Fight Against Pollution*

Summarized by Martha Gerrard

A 5-day symposium was held in Salzburg, Austria, Nov. 26–30, 1970, on the use of nuclear techniques for measurement and control of environmental pollution. More than 170 experts from 32 countries participated in the meeting, which was sponsored by the International Atomic Energy Agency (IAEA). The need for pollution control is worldwide (Fig. 1).

The keynote of the symposium was set by the paper, "Isotopic Techniques in the Study and Control of Environmental Pollution," of W. E. Mott of the USAEC Division of Isotopes Development. Application of radioisotopic techniques to environmental problems began some 15 years ago, and the number of applications has increased steadily. Effort is being directed more toward understanding specific environmental problems than toward merely evaluating the potential worth of a given technique. Increasingly, such studies are being initiated and conducted by the environmentalist and the sanitary engineer rather than by the isotope technologist, as in the past.

The technology for application of neutron activation analysis to pollution-related problems is well developed and is being widely used, but the potential of neutron-induced prompt-gamma analysis has yet to be evaluated. Although radioisotopic X-ray-fluorescence analysis is relatively new and the basic techniques needed to make it a versatile and important tool for pollutant measurement do not yet exist, it should eventually have a unique and important role in measurement and control of pollution. Important benefits could accrue from the use of radioactive tracers in municipal waste-water discharge. Mott indicated, however, that without cost reduction *and a change* in waste-water management, the radiation treatment of municipal and industrial waste water will play no important role in the immediate future. He stated:

> Those skilled in the environmental disciplines will be applying, with the help and guidance of the isotope technologist, existing and established techniques to the solution of the pollution problems. Isotope techniques will be another basic tool in the work chest of the operational scientist and will continue to contribute in a very meaningful way to the control and abatement of environmental pollution in the 1970's.

Besides the techniques mentioned above, nuclear methods include electron-capture gas chromatography and mass spectrometry along with low-level radioactivity counting. Although nuclear techniques generally cost more than do the well-established analytical methods, they have the advantages of sensitivity, selectivity, and reliability. Knut Ljunggren of Isotope Techniques Laboratory, Stockholm, pointed out that the quality of the result must be taken into account in considering the cost. Further, nuclear techniques can be used to trace pollutants back to their sources, and this could pave the way to effective control of environmental pollution.

Mercury has recently come to be considered as serious an environmental hazard as carbon monoxide, sulfur dioxide, lead, and DDT and has been studied in various countries—e.g., Yugoslavia, Sweden, and The Netherlands as well as the United States. Yugoslav workers, using neutron activation analysis, measured the uptake of mercury in domestic animals and fish around a mercury mine and processing plant but found

*Based on the symposium program, preprints, and abstracts of papers, and a report on the conference in *International Atomic Energy Agency Bulletin (Austria)*, **12**(6): 21-28 (1970).

XUM

no ill effects attributable to mercury intoxication. This may be due to an accommodation to the increased levels of mercury in the environment. Swedish workers reported finding methylation of environmental mercury to form a stable combination that is much more dangerous than inorganic mercury compounds or metallic mercury. This kind of metabolism could also occur with other elements, and therefore research on arsenic, antimony, cadmium, and selenium was proposed. Metabolic conversion in organisms can be followed only by nuclear techniques, and research on metabolites using nuclear methods may lead not only to information about the behavior and transport of substances in a system but also to acquisition of fundamental knowledge about natural processes.

The question is still open as to whether pollutants should be traced by determining components present, by adding a stable tracer with subsequent activation, or by adding a radioactive tracer. All three methods were discussed at the conference. For example, Brookhaven National Laboratory has used the sulfur-isotope ratio and sulfur hexafluoride tracer simultaneously to trace sulfur oxides in the atmosphere; the Canadians reported on the use of ^{41}Ar to study stack-gas dispersion; and activable-tracer work has been done in France and Norway, among other places. The consensus seemed to be that direct analytical methods, instead of the addition of radioactive tracers, are more likely to be applied in the future for determining, for example, the source of oil discharged into waterways.

Swedish scientists reported on the use of moss as an "environmental indicator," especially for heavy metals. Moss retains these metals since there is no exchange of nourishment with the ground, and all types of analysis may be applied.

At the University of Michigan, the behavior and variation of trace elements in air particulates have been investigated for heavily industrialized and rural areas. With the use of neutron activation analysis, a marked increase in concentrations of some elements was shown near large industrial complexes. The pollution source was also traced, with the result that iron, zinc, chromium, and a number of other elements were strongly linked to steel-producing and supporting industries. The air over the ocean was sampled and analyzed by neutron, proton, and photon activation to show the presence of pollution both near shore and over the open sea.

Swedish scientists from the University of Lund labeled the oil on a ship with 60-day ^{125}I and 8-day ^{131}I to both trace the source and determine the age of oil pollution in the sea.

Vasilii Ferronsky, Director of the IAEA Division of Research and Laboratories, stated to the participants in the symposium:

> With your active contribution, I am convinced that this first international meeting on the use of nuclear techniques on pollution problems will add to our understanding of the environment and will become a landmark symbolizing one of the important uses of radioisotopes for the welfare of mankind.

Fig. 1 The scene is in France, but it could be in almost any country. Nuclear techniques can be used in the battle against pollution. (Photo: UNESCO/Bibal, courtesy of IAEA.)

SUPPLEMENTARY BIBLIOGRAPHY

1. *Radioisotope X-Ray Fluorescence Spectrometry*, Technical Reports Series No. 115, Report of a Panel Meeting, Vienna, 1968, International Atomic Energy Agency, Vienna, 1970 (STI/DOC/10/115).
2. R. E. Greene, Tracing with Activable Tracers. Part I, *Isotop. Radiat. Technol.*, **5**: 269-277 (1968); Part II, **6**: 70-76 (1968).
3. D. E. Bryan, V. P. Guinn, R. P. Hackleman, and H. R. Lukens, Nuclear Analytical Techniques for Oil-Slick Identification, this issue of *Isotop. Radiat. Technol.*
4. R. J. Pfeifer, Measurement of Sulfur Dioxide in Stack Gases, this issue of *Isotop. Radiat. Technol.*
5. P. R. Atkins, The $^{12}C/^{13}C$ Ratio as an Indicator of Air Pollution, this issue of *Isotop. Radiat. Technol.*
6. H. P. Raaen, Comparison of Radioisotopes and Fluorescent Dyes for Estuary Pollution Control, this issue of *Isotop. Radiat. Technol.*
7. P. W. Nickola, J. D. Ludwick, and J. V. Ramsdell, Jr., Use of ^{85}Kr Tracer Technique to Define Puff Dimensions and Transport Speed, this issue of *Isotop. Radiat. Technol.*
8. P. Goodman and T. Donaghue, Kryptonate-Based Instrumentation Development for Determining Automobile Exhaust Pollutants, this issue of *Isotop. Radiat. Technol.*

Activation-Analysis and Sulfur-Isotope-Ratio Techniques for Oil-Slick Identification*

Summarized by Martha Gerrard and Francis E. McKinney

Supplementary Keywords: *activation, neutron; chemistry, analytical; reactor; pollution; industry, petroleum and natural gas; industry, petroleum refining; industry, transportation; aluminum; arsenic; barium; bromine; gallium; iodine; manganese; nickel; sulfur; vanadium.*

Abstract: *In preliminary tests, trace-element contents of crude and of residual fuel oils as determined by neutron activation analysis and sulfur-isotope ratios as determined by mass spectrometry differed sufficiently from sample to sample that they could be used to differentiate among 40 oil samples. Trace-element concentrations were affected by exposure to the environment in the first hours after the oil was spilled, but they eventually stabilized.*

One of the environmental problems becoming acute in the United States is the pollution of commercial waterways—rivers, harbors, and coastal waters—by the deliberate dumping or accidental leakage or spillage of oil. Dumping is prohibited by law, and leaks or spills are required to be reported to the proper authorities, with cleanup cost to be paid by the party responsible. Violations of the law are frequent; yet currently there is no technique available for identifying the guilty party.

Gulf General Atomic, under contract to the Division of Isotopes Development, undertook to develop a method for identifying the source of an oil slick—e.g., a particular ship, drilling platform, or bunker station. The results of two methods studied are briefly reported here: (1) the trace-element content as determined by neutron activation analysis and (2) the sulfur-isotope ratio as determined by mass spectrometry. The mass-spectrometric analyses were made at Brookhaven National Laboratory.

EXPERIMENTAL PROCEDURE

Oil samples were activated in the Gulf General Atomic 250-kW TRIGA Mark I research reactor. Induced activities were measured by multichannel gamma-ray spectroscopy, the shorter lived activities being counted with a 3- by 3-in. solid NaI(Tl) crystal scintillation detector and a 400-channel pulse-height analyzer and the longer lived with a 36-cm^3 Ge(Li) semiconductor detector coupled to a 4096-channel pulse-height analyzer. The digital pulse-height data from the NaI(Tl) spectrometer were fed onto punched cards, and the Ge(Li) spectrometer data were read onto computer-compatible magnetic tape.

The effect of weathering was studied on oil samples that had been contacted with water in plastic trays equipped with an electric stirrer to simulate either

*Summary of USAEC Report GA-9889, Development of Nuclear Analytical Techniques for Oil-Slick Identification (Phase I) by D. E. Bryan, V. P. Guinn, R. P. Hackleman, and H. R. Lukens, Gulf General Atomic Incorporated, P. O. Box 608, San Diego, Calif. 92112.

XUM

gentle or vigorous wave action. The effect of bacterial action was studied with cultures grown in the presence of oil. Samples were exposed to ultraviolet light in some experiments.

Twenty crude oils and 20 residual fuel oils were selected for investigation from the many obtained from several petroleum research laboratories. These 40 samples represented a wide geographical range, both domestic and foreign. The samples were washed by a standard procedure developed to remove suspended droplets of brine or water-soluble matter.

Two samples of each oil were irradiated: A weighed 1-ml sample was irradiated for 1 min in the pneumatic-tube position of the reactor at a thermal-neutron flux of 2.5×10^{12} n/(cm^2)(sec) and counted for 1 min after a 1-min decay; a 5-ml sample was irradiated for 30 min in the rotary specimen rack at a thermal-neutron flux of 1.8×10^{12} n/(cm^2)(sec) and counted after a few hours' decay and again after 24 to 30 hr decay. Samples—13 ml—of both original oils and oils that had been exposed to simulated environmental conditions were sent to Brookhaven for mass spectrometric analysis.

RESULTS

Twenty-two elements were observed in the various samples—some occurring in all samples and some in only a few samples. Elements that occurred frequently and were considered potentially useful for identification because of their differing concentration ranges were V, Mn, Br, Ni, S, Al, I, Ga, Ba, and As. The elements Zn, In, Sn, Co, Mo, La, Sr, and Au occurred rarely, and the presence of one or more of these might give supplemental information to that obtained from the first group. Of questionable usefulness are Na, Cl, Sb, and Sm because the Na and Cl contents often increase greatly after contact with salt water, Sb is never present after exposure to brine, and the natural association of the rare earths is such that Sm cannot be expected to be a useful independent identification element.

Vanadium was selected as the most useful element because of its stability. It was the least affected by the simulated environmental exposures, being merely concentrated somewhat as a result of evaporation of the more volatile components of the oil, but the resulting concentrations were independent of the kind of exposure. The frequency distribution of vanadium detected in the samples conforms with the literature values. The frequency distribution was not altered by any of the simulated environmental exposures. These results indicate that the log normal distribution curve of vanadium concentrations may be used in a statistical analysis. Similarly, the distributions of Mn, Br, S, Al, Ni, Ga, Ba, As, and I values indicated that using log normal distributions in statistical analyses is reasonable. Nickel concentrations also were relatively independent of the kind of exposure the oils received.

After determinations of the 10 potentially useful elements, the 40 samples were grouped according to their vanadium concentrations. In the majority of cases, those samples which had the same ranges of vanadium concentrations showed sufficient variation in manganese concentrations to permit differentiation by these two elements. The 14 samples that could not be differentiated by these two elements could be differentiated on the basis of nickel, with the exception of one pair; but in this pair the concentrations of other elements were sufficiently different to permit characterization. Crudes and residuals were lumped together in this experiment.

Sulfur-Isotope Ratios

The sulfur-isotope ratios of oil reported by Brookhaven ranged from −9.9 to +16.3, following a normal distribution, and did not change appreciably during the simulated environmental exposures. The sulfur-isotope ratio of the oil in a slick therefore would be useful in some cases. However, refining does appreciably affect the ratio, and the source of crude oil probably cannot be identified from an analysis of residual oil.

USEFULNESS OF METHODS

There was some question originally as to whether the trace-element composition of an oil slick could be definitively related to point of origin of the oil. Further, variations in composition resulting from blending and handling operations magnify the task of using a computer bank of reference information for complete search and comparison operations, and computer processing of the data would be very desirable for both speed and convenience in making such analyses. However, comparison of data on crude oils from different regions suggests that some general information can be useful. For example, in Venezuelan and California crudes, concentrations of vanadium and nickel are high, while in Oklahoma, Texas, and Louisiana crudes they are low. Unfortunately, not enough data are available in the literature to allow this approach to be evaluated. A reference library of regionally typified oil data, with associated probabili-

ties for the occurrence of particular trace-element concentration patterns, could be used to narrow the field of suspects in a given incident. Analysis of an oil slick and a record of ship movement would be submitted to the computer, which would assign probabilities of the oil's regional origin.

Two different oils are unlikely to have the same concentration pattern of all easily discernible trace elements. Furthermore, since several trace elements are not influenced greatly by exposure to wave action, ultraviolet light, bacteriological attack, or elevated temperature, it is highly probable that oil samples from the same production run or mix can be identified at a high confidence level by the activation analysis technique even if one sample has been exposed to the environment.

The $^{12}C/^{13}C$ Ratio as an Indicator of Air Pollution*

By Patrick R. Atkinst

Supplementary Keywords: *ecology; material tracing; isotopic analysis; precipitation; sorption; method; theory, applied; atmosphere; comparison; isotope abundance; in situ tracer.*

Abstract: *The stable isotopes of several atmospheric constituents possibly can be used as natural tracers of pollutants and as indicators of general levels of pollution. This paper describes a recent preliminary study in which CO_2 samples were collected by freeze-out and precipitation procedures and the $^{12}C/^{13}C$ ratios of the samples were determined. The results indicate that the $^{12}C/^{13}C$ ratio in atmospheric CO_2 can provide useful information about the degree—and perhaps the sources—of pollution that affects a given area. Freeze-out sampling is less convenient but more reliable, and probably more accurate, than precipitation. With some technique development and with calibration against freeze-out results, the precipitation procedure may prove to be usable.*

Carbon dioxide is a pollutant that has aroused considerable concern in the last few years because of its potential global effects.[1–4] In addition, since CO_2 is a by-product of all combustion processes, which may account for as much as 90% of the man-made air pollution in the United States, it should be an excellent indicator of pollution levels. Previous studies of this possibility have not produced significant results because of the masking effect of natural resources and of CO_2 at ambient levels.[5] However, this difficulty can be overcome if the $^{12}C/^{13}C$ ratio is observed rather than the total amount of CO_2 in the gas.

Ambient CO_2 concentrations of about 300 to 330 ppM have been reported for many areas of the world. Some relatively scarce data indicate that the CO_2 content of the atmosphere is increasing as a result of man's activity.[4] In a given location, the CO_2 level can vary diurnally as the photosynthetic cycle is affected by sunlight and seasonally as the plant-growth cycle is affected by weather. Together, these factors make difficult the use of total CO_2 concentration as a reliable indicator of man-made air pollution.[5] Therefore, even though CO_2 appears to be a stable constituent produced in relatively large quantities by pollution-causing processes and is very easy to collect and to determine, the use of this gas as a pollution indicator has not been completely successful.

However, the observed carbon-isotope ratio apparently could be used to determine ambient pollution levels. All carbonaceous fuels have a characteristic $^{12}C/^{13}C$ ratio that results from the isotope kinetics of their formation. Plants tend to fractionate carbon[6] and to prefer the lighter isotope, ^{12}C. The degree of fractionation may be a complex function of the type of plant, growing conditions, growth rates, etc. Carbonates[7] in equilibrium with gaseous CO_2 tend to preferentially concentrate the heavier isotope, ^{13}C. When common fuels are burned, the CO_2 produced is tagged naturally by the carbon-isotope ratio of the original fuel and therefore can be monitored at distances removed from the source. The data can be used to estimate, for the area of interest, the degree of pollution that results from the source in question.

The results of the carbon-isotope-fractionation processes are evident from Fig. 1, which gives the

*Presented as Paper No. CP 37C at the Second International Clean Air Congress, Washington, D. C., December 1970.

†Environmental Health Engineering, The University of Texas at Austin, Austin, Tex. 78712.

XUM

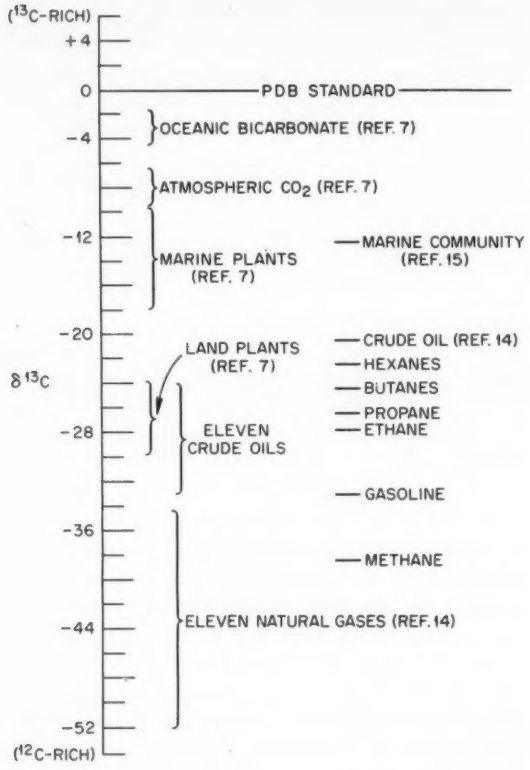

Fig. 1 Carbon reservoirs (PDB = Peedee *Belemnitella Americana* limestone).

carbon-isotope ratios of several common fuels. The changes in the carbon-isotope ratios are reported as $\delta^{13}C$, which is calculated from the equation

$$\delta^{13}C = \frac{(^{13}C/^{12}C)_{sample} - (^{13}C/^{12}C)_{standard}}{(^{13}C/^{12}C)_{standard}} \times 1000 \quad (1)$$

The $(^{13}C/^{12}C)_{standard}$ is that of a natural material, usually a limestone. In this study all the test data are reported relative to a cretaceous belemnite, *Belemnitella Americana*, from the Peedee formation of South Carolina (PDB). On this scale, a "light sample," i.e., one that contains a large amount of ^{12}C, will have a large negative number. As the ^{13}C content of the sample increases, the $\delta^{13}C$ value will approach zero. A positive $\delta^{13}C$ value indicates that a sample is considerably enriched in ^{13}C.

Carbon dioxide samples can be obtained easily and quickly by passing the air through a saturated solution of barium hydroxide, $Ba(OH)_2$. The CO_2 will react rapidly to form the stable precipitate, barium carbonate, $BaCO_3$, which can be collected by filtration and stored for future analysis. However, significant fractionation of the carbon isotopes occurs during the precipitation[8,9] and causes apparent isotopic ratios that are very different from the true values. For this reason, the more difficult and cumbersome freeze-out procedure is normally used. The air sample is slowly passed through a trap immersed in Dry Ice to remove water vapor and then through a second trap immersed in liquid nitrogen to condense the CO_2. The collected sample must be transferred to a high-vacuum flask for storage.

The objectives of the preliminary work described here were

1. To observe the magnitude of the changes that occur in the $^{12}C/^{13}C$ ratio of CO_2 samples with areas influenced by various major sources of pollution.

2. To compare the results of the freeze-out and precipitation sampling procedures.

THEORY

The carbon-isotope fractionation that occurs when CO_2 is precipitated by $Ba(OH)_2$ is not understood fully. Evidently, the fractionation results from the mass differences in the isotopes producing slightly retarded diffusion rates for ^{13}C compounds and slightly reduced impaction velocities. Baertschi[8] indicates that the fractionation is controlled by absorption rather than diffusion. In absorption, the impact velocity of the ^{13}C-compound molecules is of prime importance. In an experiment in which CO_2 and N_2 were bubbled through five gas-washing bottles arranged in series and containing 100 ml of 0.45*N* $Ba(OH)_2$, Baertschi found that the fractionation ratio was the same in each bottle, even though the CO_2 concentration decreased greatly as absorption occurred along the train. At the low flow rate of 11 cm^3/sec through the total of 500 ml of 0.45*N* $Ba(OH)_2$, the CO_2 collection efficiency was only 90%. If 100% collection could be attained, the carbon-isotope ratio of the collected sample would, of necessity, be the same as that of the CO_2 in the incoming gas. However, 100% collection apparently is difficult to accomplish.

The freeze-out procedure appears to achieve nearly 100% collection of CO_2 and to cause little fractionation of the sample. The difference between the freezing points of $^{12}CO_2$ and $^{13}CO_2$ is small enough that preferential removal is not significant. The freeze-out process does remove other substances that can con-

tribute to the mass-44 or mass-45 mass spectrometric beam. Nitrous oxide, N_2O, is a major contaminant.[9] It has molecular weights of 44, 45, and 46 and has physical properties similar to those of CO_2. Therefore, a correction must be made for the presence of this gas in freeze-out samples.

A correction must also be made to freeze-out and precipitation samples for the ^{17}O present in the CO_2 sample. The compound $^{12}C^{16}O^{17}O$ may account for as much as 6.5% of the mass-45 beam in an ambient-air sample and would normally be interpreted as $^{13}C^{16}O^{13}O$. A method for correcting the data was proposed by Craig[10] and was modified by Land[11] for the instrument used in this study.

PROCEDURES

This project was conducted in Austin and Houston, Tex., during the late fall and winter of 1969–1970. Grab samples of air were collected at several locations, and the CO_2 was removed by the precipitation and freeze-out procedures.

The air samples from which CO_2 was to be concentrated by the freeze-out procedure were collected in 5- or 19-liter flasks and were returned to the laboratory for treatment. A special vacuum manifold was used to bleed the air sample from the flask through the cold traps at reduced pressure. A calibrated manometer indicated directly the amount of CO_2 present in the sample. The condensed CO_2 was then transferred to a high-vacuum flask for storage and for mass spectrometric analysis later.

The air samples for CO_2 removal by precipitation were collected in 20-liter plastic bags, passed slowly through a gas-washer bottle that contained 100 ml of a saturated solution of $Ba(OH)_2$, and collected in a second bag. The collected effluent was then passed through the gas washer a second time and was caught again. After a third pass through the $Ba(OH)_2$ solution, the air sample was discarded. The $BaCO_3$ precipitate was filtered off through a Gelman type-A glass-fiber pad, washed with ethanol, and stored for later analysis. In the laboratory the precipitate was reacted with anhydrous phosphoric acid, and the CO_2 released was collected in a manifold by the freeze-out process.

The $^{12}C/^{13}C$ ratios in the gaseous samples of CO_2 were determined with a Nuclide model 6-60 mass spectrometer similar to that described by McKinney and associates,[12] and which has a 60° sector field and a double collector. The instrument is sensitive enough to detect accurately quantitative differences in carbon-isotope abundances of 1 part in 10 thousand.

RESULTS

Table 1 gives the data from this study. Results from the freeze-out procedure indicate that CO_2 from the air of the typical residential area in Austin, Tex., during the daylight hours has the $\delta^{13}C$ value −8.35. In the early hours before sunrise in a typical urban area, the $\delta^{13}C$ value might approach −10 to −11 owing to fractionation of CO_2 by plants and become less negative during the day.[6] The significantly more negative $\delta^{13}C$ values for samples taken late in the evening indicate that CO_2 from the combustion of "light" fuels is added to the atmosphere of the residential area during the day. The most negative $\delta^{13}C$ values were obtained for samples taken at a heavy-traffic intersection. A sample taken during a rush-hour period gave the value −20.43; during normal traffic the average value was −14.02.

The actual degree of pollution caused by automobile exhaust can be estimated from the equation

$$\frac{(CO_2)_{\text{pollutant}}}{(CO_2)_{\text{normal}}} = \frac{(\delta^{13}C)_{\text{normal}} - (\delta^{13}C)_{\text{mixture}}}{(\delta^{13}C)_{\text{mixture}} - (\delta^{13}C)_{\text{pollutant}}} \quad (2)$$

This equation was first used by Parker,[13] not in studying air pollution but in ascertaining that more than half the dissolved organic material in the water of the Houston Ship Channel is derived from pollution. Since $\delta^{13}C$ values of pure automobile exhaust, i.e., $(\delta^{13}C)_{\text{pollutant}}$, were about −30.5 and since the $\delta^{13}C$ of clean air in Austin is estimated to be −8.35, i.e., $(\delta^{13}C)_{\text{normal}}$, Eq. 2 indicates that automobile exhaust is the source of possibly 28% of the total CO_2 in the atmosphere near normal traffic. The very-heavy-traffic $\delta^{13}C$ (−20.43) indicates that 55% of the total CO_2 was produced by automobile exhaust.

Air samples taken inside a brewery at Houston show that the CO_2 present within the building has a very negative $\delta^{13}C$ value, i.e., the CO_2 is rich in ^{12}C. This result is to be expected since much of the CO_2 present is probably produced by the fermentation process.

The downwind $\delta^{13}C$ values near an oil refinery indicate a high degree of pollution. Crude oils, gaseous derivatives, and natural gases have been shown[14] to have $\delta^{13}C$ values of −47 to −21. The freeze-out procedure gave an average downwind $\delta^{13}C$ value of −15.90. If −8.35 is assumed to be the $\delta^{13}C$ value for clean air, Eq. 2 indicates that ~29% of the downwind total CO_2 was produced by the oil refinery.

The $\delta^{13}C$ data from a steel-mill plume gave little information about the plume's chemical character. The

XUM

Table 1 CO_2 Content and $\delta^{13}C$ Values for Air Samples

Sample location	Time of day	Freeze-out procedure		Precipitation procedure	
		CO_2, %	$\delta^{13}C$	CO_2, %	$\delta^{13}C$
		Nov. 12, 1969			
Residential	10 p.m.	0.0374	−13.98	–	–
		Nov. 13, 1969			
Residential	8 p.m.	0.0284	−15.27	0.005	− 5.58
Heavy traffic		0.0741	−20.43	–	–
		Nov. 14, 1969			
Residential	10 a.m.	0.0305	− 9.33	0.0100	−12.18
		–	− 8.29	–	–
		–	− 8.21	–	–
Moderate traffic	8 p.m.	0.0536	−13.23	0.0375	− 7.61
		–	−14.63	–	–
		0.0431	−14.30	0.0360	− 9.88
		0.0274	−13.92	0.0125	−10.67
		Nov. 20, 1969			
Moderate traffic	6 p.m.	0.0320	−14.71	0.0550	− 9.54
		0.0280	−14.52	0.0325	− 9.44
		–	–	0.0335	− 9.72
		Dec. 2, 1969			
Brewery interior		0.0320	−22.70	0.0085	−10.23
Oil refinery (downwind)		0.0360	−16.20	0.0035	− 9.98
		0.0326	−15.70	–	–
Steel mill (downwind)		0.0305	− 9.75	–	–
		–	− 9.85	–	–

$\delta^{13}C$ values for coal[7] were in the range −28 to −22. The average freeze-out $\delta^{13}C$ value for gas directly in the plume and ~1000 ft downwind from the source was −9.8. Thus, even though the mill was using coke as a heat source, the gas was much richer in ^{13}C than was expected. Perhaps the limestone flux used in the furnaces contributed large amounts of "heavy" CO_2.

Comparison of the freeze-out data with the precipitation data (Table 1) shows that, in all cases where similar samples were collected, the samples isolated by the precipitation procedure were enriched in ^{13}C relative to the freeze-out samples, except the residential-air sample collected at 10 a.m. on November 14. The CO_2 concentrations determined from these precipitation samples were consistently low, often less than half the normal ambient values. This result possibly indicates that the precipitation system was leaking or that precipitation efficiencies were extremely low. The former explanation is the more logical.

Enrichment of the precipitation samples in ^{13}C was not expected. Since ^{12}C is used preferentially in the fractionation that occurs in the precipitation process, the $\delta^{13}C$ values should have been more negative than those found by the freeze-out procedure.[8] A possible explanation for this anomaly lies in the carbonate–CO_2 equilibrium. It has been reported[7] that, in a general way, oxidized carbon compounds tend to concentrate ^{13}C and reduced compounds to concentrate ^{12}C. For example, carbonates tend to exchange ^{12}C-compound molecules for ^{13}C-compound molecules in an equilibrium reaction. When CO_2-containing gas is bubbled slowly through a medium that contains $BaCO_3$ precipitate, suspended by the agitation of the bubbles, the ^{12}C–^{13}C exchange process may occur. The measured $\delta^{13}C$ values will then become less negative. The extent of this exchange reaction will be time dependent.

Therefore, for several reasons, the $\delta^{13}C$ value obtained by a precipitation technique will depend on

the rate of flow of gas through the absorbing solution. It should be possible to develop a standardized precipitation technique and to calibrate the results of this technique against the freeze-out method so that $\delta^{13}C$ data can be used to estimate the degree of pollution from combustion sources. The development of such a procedure will be attempted at The University of Texas at Austin.

This study was limited to the stable isotopes of carbon, but other airborne pollutants of interest could also be similarly studied. For example, nitrogen, oxygen, and sulfur each have two or more stable isotopes that are normally present in measurable quantities. Measurements of these isotopes could give information about the sources of sulfur oxides and nitrogen oxides present in polluted air. The sampling procedures and data-interpretation methods will be more complex, but such studies should prove rewarding. (HPR)

REFERENCES

References marked with an asterisk have not been verified by the editors; for further explanation, see the Foreword.

1. G. N. Plass, Effect of Carbon Dioxide Variations on Climate, *Amer. J. Phys.*, **24**: 376-387 (1956).
2. F. Möller, On the Influence of Changes in the CO_2 Concentration in Air on the Radiation Balance of the Earth's Surface and on the Climate, *J. Geophys. Res.*, **68**: 3877-3886 (1963).
3. J. M. Mitchell, Jr., Recent Secular Changes of Global Temperature, *Ann. N. Y. Acad. Sci.*, **95**: 235-250 (1961).
4. E. K. Peterson, Carbon Dioxide Affects Global Ecology, *Environ. Sci. Technol.*, 3(11): 1162-1169 (1969).
5. J. F. Clarke and R. B. Faoro, An Evaluation of CO_2 Measurements as an Indicator of Air Pollution, *J. Air Pollut. Contr. Ass.*, **16**(4): 212-218 (1966).
6. C. D. Keeling, A Mechanism for Cyclic Enrichment of Carbon-12 by Terrestrial Plants, *Geochim. Cosmochim. Acta*, **24**: 299-313 (1961).
7. H. Craig, The Geochemistry of the Stable Carbon Isotopes, *Geochim. Cosmochim. Acta*, **3**: 53-92 (1953).
8. P. Baertschi, The Fractionation of Carbon Isotopes by Absorption of Carbon Dioxide (in German), *Helv. Chim. Acta*, **35**: 1030-1036 (1952).
9. H. Craig and C. D. Keeling, The Effects of Atmospheric NO_2 on the Measured Isotopic Composition of Atmospheric CO_2, *Geochim. Cosmochim. Acta*, **27**: 549-551 (1963).
10. H. Craig, Isotopic Standards for Carbon and Oxygen and Correction Factors for Mass-Spectrometric Analysis of Carbon Dioxide, *Geochim. Cosmochim. Acta*, **12**: 133-149 (1957).
11. *L. S. Land, Department of Geology, University of Texas at Austin, 78712, unpublished.
12. C. R. McKinney, J. M. McCrea, S. Epstein, H. A. Allen, and H. C. Urey, Improvements in Mass Spectrometers for the Measurement of Small Differences in Isotope Abundance Ratios, *Rev. Sci. Instrum.*, **21**: 724-730 (1950).
13. *P. L. Parker, Stable Carbon Isotope Ratio Variations in Biological Systems, in *Proceedings of the Symposium on Organic Matter in Natural Waters*, D. Hood (Ed.), Institute of Marine Science, University of Alaska, 1970.
14. S. R. Silverman, Carbon Isotopic Evidence for the Role of Lipids in Petroleum Formation, *J. Amer. Oil Chem. Soc.*, **44**: 691-695 (1967).
15. P. L. Parker, The Biogeochemistry of the Stable Isotopes of Carbon in a Marine Bay, *Geochim. Cosmochim. Acta*, **28**: 155-164 (1964).

Comparison of Radioisotopes and Fluorescent Dyes for Estuary Pollution-Control Studies*

Summarized by Helen P. Raaen

*Quirk, Lawler & Matusky Engineers, Utility of Radioisotope Methodology in Estuary Pollution Control Studies. Part I. Evaluation of the Use of Radioisotopes and Fluorescent Dyes for Determining Longitudinal Dispersion Coefficient in Estuaries, August 1969; report to the U. S. Atomic Energy Commission, Division of Isotopes Development.

Supplementary Keywords: *hydrology; ecology; material tracing; review, technical; economics; bibliography; data handling; data compilation; pollution; sea.*

Abstract: *In a series of laboratory studies, radioisotopes were compared with fluorescent dyes for use in studying estuary*

XUM

pollution control. The comparison was made by Quirk, Lawler & Matusky Engineers under a Division of Isotopes Development contract. Their report of the work is summarized here.

Knowledge of the capacity of a waterway to assimilate wastes is essential to an effective water-pollution-control program. This capacity depends on channel geometry, freshwater advection, atmospheric reaeration, dispersion characteristics, and biochemical oxidation of the wastes. In freshwater rivers and streams, these parameters are readily determined from known or measurable physical and hydraulic characteristics. In tidal waterways and estuaries, they are more difficult to measure directly.

Existing mathematical models do not permit precise descriptions of some of these parameters for estuaries. Consequently, a reliable method of determining the unknown parameters is the direct experimental measurement of the distribution of a tracer. The general procedure is to introduce into the flow a known amount of soluble tracer whose concentration can be measured at various times and places. Therefore, the selection of a suitable tracer and tracing technique is the first step to any comprehensive study of receiving waters.

Possible tracers include naturally occurring substances (e.g., salt) and added materials (e.g., dyes and sugar). The use of most of these in the field is seriously affected by variable tracer characteristics, by losses due to various combinations of decay mechanisms, and by the deleterious effects on receiving waters. Radioisotopes have several unique characteristics that make their use as field tracers attractive from a practical standpoint.

Studies were made by Quirk, Lawler & Matusky Engineers to evaluate the relative merits of dyes and radiotracers for determining certain mass-transport parameters in estuaries, as typified by the Hudson River, and to delineate areas in which radioisotopes would either provide the only practical means of estimating these parameters or appear to have significant advantages.

DISCUSSION

Scope

The studies consisted in

1. Review of the literature to locate information on existing techniques for tracer studies in waterways and on applications of dyes and radioisotopes in general hydrological investigations. (Seventy-three literature references and 28 previous tracer studies of waterways are cited.)

2. Evaluation of dye studies made previously in several selected estuaries.

3. Comparison of radioisotopes and fluorescent dyes, including a laboratory evaluation of their characteristics and behaviors in waters of various qualities.

4. Evaluation of the types of field investigations required to confirm the potential of radiotracers for dispersion measurements.

5. Review of licensing requirements and procedures for the use of radiotracers in estuaries and in hydraulic models.

Selection of Radioisotopes and Dyes

More than 100 commercially available radioisotopes and fluorescent dyes were considered. Their evaluation was based on their chemical and physical characteristics as tracers; effects of water quality on their determinations in trace concentrations; measurement sensitivities and required detection equipment; availability in sufficient variety and quantity; safety hazards; ecological effects; and costs to purchase, handle, and detect.

The 4 dyes and 14 radioisotopes selected as being suitable for use as field tracers for dispersion measurements are:

Dyes: Rhodamine B, Sulpho Rhodamine B (Pontacyl Brilliant Pink), Rodamine WT, Fluorescein

Radioisotopes: ^{3}H, ^{24}Na, ^{42}K, ^{46}Sc, ^{51}Cr, ^{58}Co, ^{82}Br, ^{85}Sr, ^{86}Rb, ^{99}Mo, ^{103}Ru, ^{122}Sb, ^{124}Sb, ^{140}Ba

Laboratory Evaluation of Certain Radioisotopes and Dyes

Of the selected potential tracers, the following were evaluated in the laboratory: Rhodamine B, Sulpho Rhodamine B, Rhodamine WT, ^{51}Cr, ^{82}Br, ^{85}Sr, and ^{124}Sb. Their behaviors in estuarine waters of various qualities were studied. It was concluded that laboratory-determined dye losses represent an upper limit and must be reduced for field application and that ^{82}Br, ^{85}Sr, and ^{124}Sb can be expected to perform properly as tracers for estuarine water bodies.

Economics

Conclusions reached regarding the economics of radioisotopes relative to fluorescent dyes as estuarine tracers are:

1. The cost of radiotracer material is generally less than the cost of dye material. The quantity ratio of required dye (kg) to radioisotope (Ci), however, may range from 0.1 to 1000 kg/Ci depending on survey conditions, tracer decay rate, and minimum detectable concentration.

2. In most practical cases, the overall cost of the required radioisotope-survey equipment is only 5 to 10% higher than the cost of the dye-survey equipment. For example, a fully equipped scintillation counter costs ~$2500 compared with $2000 for a Turner model III fluorometer. In general, the equipment required for a radiotracer survey includes most of the dye-survey equipment plus a few additional instruments.

3. Manpower cost for radioisotope surveys is significantly higher than for dye surveys. The higher cost results from the expense of a license, public relations, safety precautions, and the required high level of personnel training.

4. Initial large-scale radiotracer surveys may cost one to two times as much as a dye survey. However, in repeated runs radiotracers may be as economical as dyes. Experience with regulatory matters and nontechnical aspects significantly reduces the overall cost.

5. The overall cost of a small-scale or a hydraulic-model radiotracer experiment is essentially the same as that of a dye study. Furthermore, few, if any, public-relations, safety-precaution, and licensing problems are encountered in such surveys.

6. The extra cost of initial radiotracer applications must be evaluated on the basis of the need for and importance of a more refined measurement, as well as on the availability and reliability of other approaches.

Mathematical Models

Currently available mathematical models that describe the assimilation capacity of estuaries are not very sensitive to variation in the longitudinal dispersion coefficient. Therefore, an added refinement in dispersion measurements is of limited importance in this regard.

Precise longitudinal dispersion measurements are necessary in several areas because of the high sensitivity of the mathematical models employed. These areas include the prediction of the extent of salt intrusion in estuaries, thermal-pollution problems, and waste-diffuser studies. In these studies, radiotracers may find their greatest utility.

The 18 potential tracers were compared on the basis of selected mathematical models that consider tracer characteristics and behavior in the field. Several comparative aspects were expressed in terms of the cost of required materials, survey equipment, and manpower needs. The influence of those field-study problems that cannot be expressed in monetary terms was also considered. Furthermore, the comparison considered current and future research and development in addition to project cost patterns of tracer technology.

A generalized solution of the selected mathematical model was developed to make possible the selection of the best tracer, the survey duration, and the required optimum tracer quantity. This solution is valid in one-dimensional waterways, such as rivers, streams, tributaries, and estuaries.

Areas of Advantage for Radioisotopes

Radiotracers appear to offer significant advantages in some areas, which include:

1. Areas where the use of fluorescent dyes is impractical, such as (*a*) areas near industries that discharge thermal wastes or wastes containing dyes or chemical oxidants and (*b*) large bodies of water where the dilution is great.

2. Estuarine waters, when algae, suspended solids, turbidity, and natural organic components of seawater are present. In these cases, the use of radiotracers results in more-reliable measurements.

3. Hydraulic-model runs. Because of the release of extremely low quantities of radioactivity in a highly controlled and enclosed system, the associated safety, handling, and public-relations problems are minor.

4. Measurements of atmospheric reaeration, heat-transfer coefficients, freshwater flow and its attenuation, subsurface water inflow and outflow, and identification of pollution contributors and dredging spoil.

SUMMARY

The results of this comparison are summarized in Tables 1 and 2, which are, respectively, a priority list of tracers arranged according to suitability of use and a list of the advantages and disadvantages of both radioisotopes and fluorescent dyes as tracers in estuary-pollution studies.

XUM

Table 1 Priority List of Tracers Suitable for Prototype and Hydraulic Models*

Prototype and large-scale surveys	Chemical form	Hydraulic-model and small-scale surveys	Chemical form
^{46}Sc	$ScCl_3$	Rhodamine B	
^{124}Sb	$SbCl_3$	^{82}Br	NH_4Br
Rhodamine WT		^{24}Na	$NaCl; NaNO_3$
^{122}Sb	$SbCl_3$	^{42}K	$KCl; K_2CO_3$
Sulpho Rhodamine B		Fluorescein	
^{103}Ru	$RuCl_4 \cdot 5H_2O$		
^{99}Mo	$(NH_4)_2MoO_4$		
^{140}Ba	$BaCl_2$		
^{58}Co	$CoCl_2$		
^{51}Cr	$Cr(NO_3)_3 \cdot 9H_2O$		
^{85}Sr	$SrCl_2 \cdot 4H_2O$		

*Obvious deletions include such isotopes as ^{82}Br, whose half-life is too short for extended studies, and ^{3}H, because it is a beta emitter.

Table 2 Summary of Comparison of Radioisotopes and Fluorescent Dyes for Estuary Pollution Studies

Advantages	Disadvantages
Radioisotopes	
1. Maximum sensitivity and specificity	1. Approval and licensing required from governmental agencies
2. Chemical and physical stability	2. Possible adverse public reaction
3. Known and constant decay rate	3. Possible health hazards and disastrous effects of an accident
4. Freedom from natural interferences	4. Scarcity of persons experienced in radioisotope technology
5. Possibility of multitracer studies	5. Expense of a license, safety precautions, and nontechnical aspects; these may offset the low material cost
6. Ability to limit to selected reaches the travel and the dispersion pattern of tracer	6. Possible delay of up to several months before production and delivery of a radioisotope in curie amounts
7. Ability to use a tracer with the specific characteristics (e.g., decay rate) of the pollutant under study	
8. Constancy of characteristics, which eliminates the tracer as a variable when the results are generalized	
Fluorescent Dyes	
1. Relative speed	1. Ill-defined decay rate that depends on the characteristics of the environment
2. Ease of handling	2. Interferences in measurement due to presence of algae, turbidity, etc.
3. Minimum health hazard and no public concern	3. Low sensitivity at low concentration
4. Immediate availability	4. Instability in the presence of chemical oxidants and of dissolved and suspended solids
5. Adequacy of untrained technologists	5. Temperature sensitivity, which requires accurate temperature measurements in areas where longitudinal and lateral temperatures vary markedly
6. Visual tracings	
7. Possible cheapness	

Tritiated-Water-Vapor Method for Studying Lake Evaporation*

By P. J. Barry† and W. F. Merritt†

Abstract: *A method for using tritiated water vapor for the study of evaporation mechanisms in a natural setting is described.*

The primary purpose of the Perch Lake Project at Chalk River Nuclear Laboratories (CRNL), being carried out with the cooperation of the Inland Waters Branch of the Department of Energy, Mines and Resources of the Government of Canada and the Hydrometeorology section of the Department of Transport, is to exploit the use of radioactive tracers in the study of all aspects of the hydrological cycle in a small, well-defined basin. Within this broad objective, a number of individual studies are being conducted, one of which—the use of tritiated water vapor in studying evaporation mechanisms in a natural setting—is believed to be unique.

Perch Lake, a small (0.54 km^2) lake on the CRNL property, is typical of thousands of lakes in eastern Canada. It differs from the others, however, in one important feature: Its water contains about 400 pCi/ml of tritium coming from the liquid-disposal area, where waste solutions containing several thousand curies of tritium (as HTO) are discharged annually. From pits in the ground, the liquid moves in the surface- and groundwaters of Perch Lake swamp into the lake, so that the groundwater forms a ^3H reservoir estimated at about 8000 Ci. The lake itself is located at the lower end of a well-defined drainage basin, occupying a saucer-shaped bedrock depression. One small creek provides the only outlet for surface water for the whole basin. The lake, which has an average depth of ~2 m, is frozen from the end of November to early April, and the margins are heavily vegetated in summer. The winds are markedly channeled by the valley of the Ottawa River, and 70% of all winds blow either northwest or southeast.

The method being used is described here.

*Summary of "Perch Lake Evaporation Study," Canadian Report AECL-3557, 1970; also in *Isotope Hydrology 1970*, Symposium Proceedings, Vienna, 1970, pp. 139-151, International Atomic Energy Agency, Vienna, 1970 (STI/PUB/255).

†Biology and Health Physics Division, Chalk River Nuclear Laboratories, Atomic Energy of Canada Limited, Chalk River, Ontario.

PRINCIPLES OF METHOD

Consider a surface oriented vertically and normal to the mean wind direction in the air above the downwind shore of the lake and having a height dz at elevation z and width dy at position y, measured normal to the mean direction of the wind. Let $c(y,z,t)$ be the air concentration of tritium and $u(y,z,t)$ the speed of the air passing through the surface at time t. Then the amount of tritium passing through the surface in the time interval dt centered at t is

$$A(y,z,t) = c(y,z,t)\,u(y,z,t)\,dy\,dz\,dt \qquad (1)$$

and the amount passing through an unlimited vertical strip of width dy is

$$A(y,t) = dy\,dt \int_0 c(y,z,t)\,u(y,z,t)\,dz \qquad (2)$$

The amount passing through the vertical strip in the finite sampling interval τ to θ is

$$A(y,\theta-\tau) = dy \int_\tau^\theta \int_0^\infty c(y,z,t)\,u(y,z,t)\,dz\,dt \qquad (3)$$

and the total amount of tritium in the air passing over the end of the lake is

$$A(\theta-\tau) = \int_{-\infty}^{\infty} \int_\tau^\theta \int_0^\infty c(y,z,t)\,u(y,z,t)\,dz\,dt\,dy \qquad (4)$$

If the air approaching the upwind shore of the lake is free of tritium, the amount $A(\theta-\tau)$ may be equated to the amount of tritium coming from the lake during the time interval τ to θ. The experiments are carried out when the wind direction is such that this condition is fulfilled. The very small concentration of tritium in the atmosphere from cosmic and nuclear weapon sources is not detectable by the method used to assay the tritium content of the air samples.

Assuming, then, that the wind field can be adequately represented as a scalar quantity, Eq. 4 allows the tritium evaporation to be determined accurately from measurements of c and u. In practice, a number

XUM

of additional assumptions must be introduced, the most important of which are:

1. The maximum height of sampling is necessarily restricted, although it should be possible to select a height h at which $c(h) \approx 0$.

2. The concentration of tritium can be measured only over relatively long time periods; therefore the product $u(t)\,c(t)$ must be approximated by the product of the individual means, $\bar{u}\bar{c}$. Since the instantaneous concentrations and wind speeds are the instantaneous departures from their means, $c(t) = \bar{c} + c'(t)$, $u(t) = \bar{u} + u'(t)$, the product $u(t)\,c(t)$ is $u(t)\,c(t) = \bar{u}\,\bar{c} + \bar{c}\,u'(t) + \bar{u}c'(t)\,u'(t)\,c'(t)$ and the time integral over the sampling interval τ to θ, where $\theta - \tau = s$, is

$$\int_\tau^\theta u(t)\,c(t)\,dt = \bar{u}\,\bar{c}\,s + \int_\tau^\theta u'(t)\,c'(t)\,dt \qquad (5)$$

(The terms $\int_\tau^\theta u'(t)\,dt$ and $\int_\tau^\theta c'(t)\,dt$ are, by definition, zero.)

The magnitude of the error introduced by the approximations depends on the relative magnitudes of the two terms on the right-hand side of Eq. 5. Unfortunately, there is no way of estimating the magnitude of the second term directly, but its relative importance can be inferred from successive, very-short-period measurements of wind speed and humidity, using techniques developed for the eddy-correlation method of measuring evaporation. The error is held to a minimum by using the shortest possible averaging times (30 to 60 min), thereby avoiding at least the effects of long-period components of turbulence.

3. With the present experimental arrangement, c and u can be measured at two shore-line stations (on opposite sides of the lake), but it is impossible to estimate how the term A varies in the y (crosswind) direction. At Perch Lake this restriction means that the measured values of c and u can be used only when the mean wind is within an angle of 50°, or less if there are large components of crosswind turbulence having time periods of the same order of magnitude as the averaging times. Therefore, tritium evaporation can be measured only during limited time periods and over a restricted area of the lake surface. However, this is sufficient for many experimental purposes and, within these limited objectives, the errors introduced by assuming that $dA/dy \approx 0$ are acceptable.

Introducing these simplifications, Eq. 3 takes the practical form

$$\overline{E(\Delta y,s)} = \Delta y \int^h \overline{c(z)}\,\overline{u(z)}\,dz \qquad (6)$$

where $\overline{E(\Delta y,s)}$ is an estimate of $A(y,s)$ along a strip Δy wide and averaged over the sampling interval s; h is the height of the sampling tower; and Δy is the width of the air column normal to the mean wind direction for which the measured values of u and c are representative.

If L is the length of the lake in the mean wind direction at the point y, the evaporative flux, F, averaged over time s and area $L\,\Delta y$ is

$$F(\Delta y,s,L) = \frac{\overline{E(\Delta y,s)}}{L\,\Delta y} = \frac{1}{L}\int_0^h \overline{c(z)}\,\overline{u(z)}\,dz \qquad (7)$$

The net upward flux of tritium decreases with distance of travel as its concentration in the vapor builds up in the boundary layer. The rate of change of flux with distance decreases progressively as the conditions in the lower part of the boundary layer approximate a steady state. For most practical purposes, this steady state may be sufficiently well approached after a reasonably short travel distance. However, neither the minimum distance needed to establish steady state nor how this is affected by meteorological factors is known. Most commonly used evaporation equations based on aerodynamic models assume that a steady state exists in the boundary layer, and it is important to find out the minimum fetch to which they can be applied. For example, Marciano and Harbeck[1] tested several evaporation equations at Lake Hefner and found that, of the two which agreed most closely with the daily evaporation, one assumed local evaporation to be variable with length of wind sweep while the other assumed it to be constant.

EXPERIMENTAL PROCEDURE

One of the first Perch Lake studies will determine the average flux of tritiated water vapor as a function of the fetch using Eq. 7. Two sampling towers have been placed over the lake on a line between the two shore towers, the line lying in the direction of the most frequent winds. Each is out from the shore a distance equal to one-third the total length of the lake. With this arrangement, $F(\Delta y,sL)$ for L equal to about 250, 500, and 750 m can be estimated.

A measured volume of air is drawn through a bed of Molecular Sieves type 4A, which removes the water vapor. Stainless-steel vessels containing 100 g of sieves, and able to trap up to 10 g of water, are equipped with vacuum-tight valves on both inlet and outlet tubes. The air sampling rate is adjusted so that about 5 g of water is collected during the sampling period. The water is

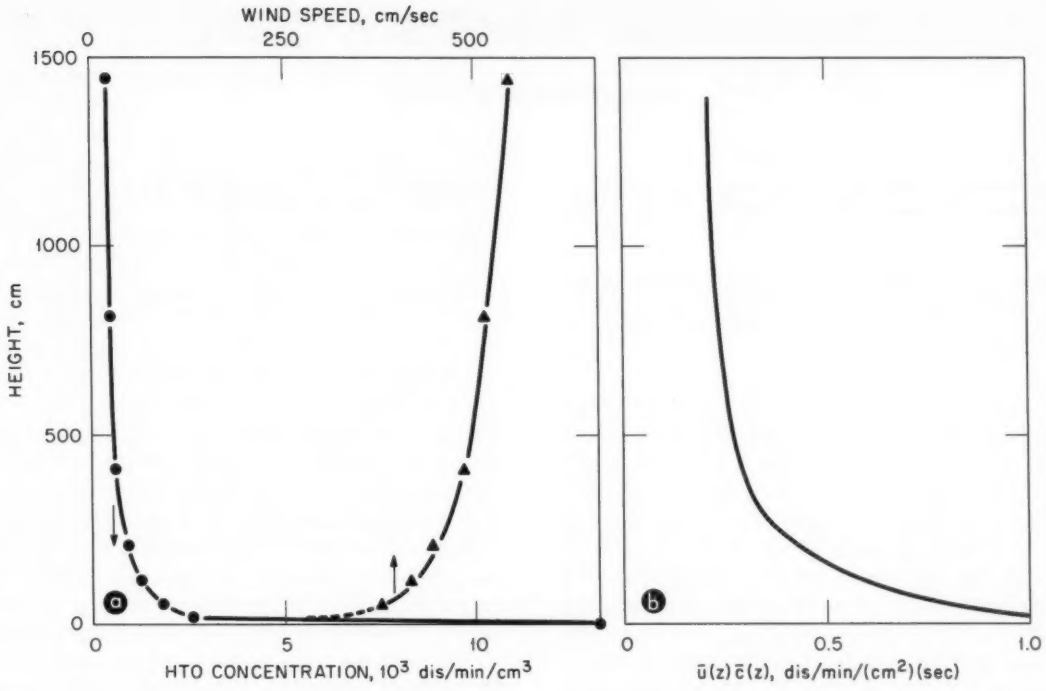

Fig. 1 Typical data on variation of (*a*) wind speed and tritiated-water-vapor concentration and (*b*) HTO concentration in air with height above the lake.

distilled for 5 hr at 54°C, under high vacuum, into a weighed glass trap cooled in liquid N_2. The trap is again weighed, and the amount of water collected is obtained by difference. The tritium content is measured by liquid scintillation counting, using standard techniques. From the amount of air sampled and the volume of water collected, the concentration of HTO in the air is calculated. Twenty-four samples can be processed per day. The sensitivity of the system is such that water containing less than 4 pCi/ml HTO can be assayed without difficulty.

The results of 28 shakedown runs (Fig. 1) indicate that the sampling and analytical systems operate satisfactorily. No difficulties are expected in actual measurement runs.

REFERENCE

1. J. J. Marciano and G. E. Harbeck, Water Loss Investigations, Lake Hefner Studies Technical Report, Geological Survey Professional Paper No. 269, U. S. Geological Survey, Washington, 1954.

XUM

Radioisotopic Sand-Tracer Study*

Summarized by Martha Gerrard

Supplementary Keywords: *material tracing; sedimentology; oceanology; preparation, labeled material;* ^{133}Xe; ^{198}Au; ^{199}Au.

Abstract: *Sand-labeling methods and sand-injection devices developed for tracing sediment movement on a dry beach, in a breaker zone, and in deep water and for field tests on the survey system are summarized briefly.*

The 3-year Radioisotopic Sand Tracer (RIST) investigation of littoral transport around Point Conception, Calif., was started in 1966. This is a location on the California coast where currents make an ~90° turn. The project objectives were to trace the movement of labeled sand in this zone after developing methods for labeling the sand with suitable radioisotopes and equipment for detecting the tracers in beach and marine environments to depths of 100 ft. In this article, the tracer preparation and injection device are described and the field tests through February 1969 are summarized. A report on the detection equipment is found elsewhere in this issue of *Isotopes and Radiation Technology.*[1]

SAND LABELING

Several radioisotopes were considered for the RIST tests, and two were actually used—^{133}Xe (0.08-MeV gamma) and $^{198,199}Au$ (^{198}Au, 0.4-MeV gamma). In field tests, the gold, because of its higher energy radiation and ability to tag sand to a higher activity level, was found to be more useful in rapidly moving systems, and it was adopted for further development of the process.

Since it had been decided to use actual sand from the Point Conception site to avoid any possible differences in hydrological behavior of the tracer from that of the sand being tested, the labeling procedure had to be one that did not change the properties of the sand. The procedure developed at Oak Ridge National Laboratory[2] for this purpose is a direct reduction of ^{198}Au chloride on the sand. The technique simulates mass labeling, i.e., labeling in which the amount of label on a particle is proportional to the weight of the particle. The sand is first fractionated by particle size, and to each fraction is added an amount of $^{198,199}Au$ that is the same proportion of the total activity to be used as that fraction of sand is to the total batch of sand being labeled. Prior to the labeling, the sand is acid washed to remove carbonate and is then treated with *p*-isopropylbenzaldehyde to provide a reducing surface, and the gold chloride is added. After 30 min, the solution is decanted, and the sand is dried and heated to 1000°C. Other methods investigated were indirect deposition of the gold by first depositing silver on the sand and replacing it by gold and both direct and indirect deposition of gold using $SnCl_2$ as the reductant. However, the method finally adopted was direct *p*-isopropylbenzaldehyde reduction, which gave a product with the best resistance to salt-water leaching and abrasion.

Sand labeled[3] with ^{133}Xe was used in some of the earlier work. This is a surface-labeling technique in which the sand is cleaned, e.g., with HCl, and then heated to 860–1100°C in a ^{133}Xe atmosphere. In tests on the labeled product, no xenon was lost by leaching in seawater. However, specific minerals in sand were found not to be labeled with the same efficiency, causing the individual grains to incorporate different quantities of ^{133}Xe adsorbed.

Labeling with ^{140}Ba–^{140}La was also considered.[4] However, the labeling procedure coats the sand grains with sodium silicate containing the radionuclide, and it was originally feared that the hydraulic properties of the sand would be altered by the treatment. Therefore this label was not adopted for the project.

INJECTION DEVICE

The most satisfactory system developed for emplacing the labeled sand is to package it in water-soluble plastic bags. About 15 g is contained in each bag. These bags of labeled sand are dropped from a shielded cylinder by a simple push-rod mechanism. The

*A joint project of the U. S. Army Coastal Engineering Research Center and the USAEC Division of Isotopes Development, with participation of Oak Ridge National Laboratory, U. S. Navy Pacific Missile Range, U. S. Air Force First Strategic Aerospace Division, U. S. Army Engineer District Office in Los Angeles, U. S. Army Mobility Command, National Aeronautics and Space Administration, and the State of California Division of Water Resources. F. N. Case, one of the project leaders for this study, kindly reviewed this article before publication.

package dissolves in 20 to 30 sec and can reach a depth of 25 ft before the bag dissolves and releases the labeled sand. For greater depths, either the packages are weighted with lead pellets or thicker plastic bags are used. The packages are delivered to the equipment ready to use and can be handled by personnel much more easily than can material that must be transferred on the site.

One of the earlier injection systems was a cylindrical hopper for the labeled sand, with attached pump and hoses through which the sand was passed.[5] A clamshell device was used next, suspended with the hinge upward so when it reached the ocean floor the sand would drop out as the container was opened by a spring mechanism. With a capacity of 40 liters, it was useful for placing sand as a point source in water seaward of the breaker zone. However, on two of three occasions on which it was used, scuba divers had to manipulate the jaws of the clamshell in order to open them completely.

FIELD TESTS[4,5]

In April 1967, procedure operation and equipment performance were tested at Cape Kennedy, Fla. The environmental conditions at Cape Kennedy are relatively mild, and necessary adjustments could be made more easily than at Point Conception. The tracer was quartz sand from California labeled with ^{133}Xe, but differences between this sediment and the local sediment were felt to be unimportant at this stage of the project. The detector system performed satisfactorily, with ^{133}Xe counts several orders of magnitude above background being obtained.

The next test was in June 1967, at Surf, Calif. (Fig. 1), where environmental conditions are somewhat harsher than at Cape Kennedy but still milder than at Point Conception. The detector system again operated satisfactorily, and it was possible to observe sediment movement for several days. Because of difficulties with the injection system, the tracer was injected into the water column, but the injection point was poorly defined.

Tests were then made in the Point Conception–Government Point area where there is a sharp change in the orientation of the California coastline. Both points are rocky promontories, and there are bluffs and extensive rocky areas on the sea floor.

Following preliminary operations in November 1967, tracer injections were made on December 1, 2, 7, and 10:

Injection area	Injection depth, ft	Labeled sand, liters	Total ^{133}Xe activity, mCi
A	30	40	1200
C	30	40	800
B	30	40	600
A	Surf zone	1	120

At areas A, B, and C (Fig. 1), average mean sand-particle sizes are 0.17, 0.175, and 0.203 mm, respectively.

Too few data were obtained to show definitely whether sand moves around Point Conception. The difficulties may have been due to too slow dispersion combined with too widely spaced tracking, too rapid dispersion of tracer to a concentration beyond the limits of detection (1 $\mu Ci/ft^2$), or burial of the tracer. However, slow dispersion was thought to be the reason for nondetection of the tracer beyond the injection point. Bouncing of the detector unit as it was towed over rock outcrops caused gain shift and noise.

To determine the effects of high- and low-specific-activity sand on the results, in May 1968 tracing tests were made in a 68-ft-long by 10-ft-wide flume con-

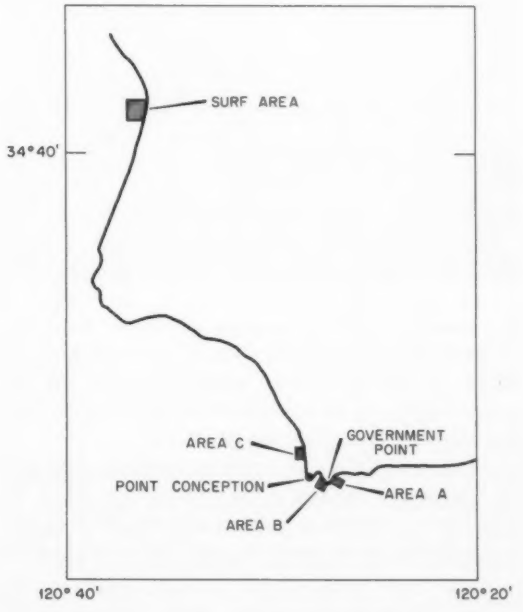

Fig. 1 Chart of Point Arguello–Point Conception area, showing location of test sites.

XUM

structed in a test basin, with a simulated beach where the sand particle size averaged 0.27 mm. The high- and low-activity ^{133}Xe-labeled tracers had specific activities of 520 and 5.28 μCi/cm^3, respectively. The results with a small amount of high-activity sand were not significantly different from those with large amounts of low-activity sand.

The next on-site test at Surf was in September 1968. Thirty-six 1-liter packets (soluble plastic) of ^{133}Xe-labeled sand (42.3 μCi/cm^3) were placed in a 180-m-long line perpendicular to the wave front, extending into the surf from a point on the beach 6 ft beyond the breaker zone. Again, paucity of data obviated any conclusions on sediment dispersal. Some burial was indicated, but complete burial seemed unlikely.

In October 1968, tests were made again at Surf, this time with ^{198,199}Au-labeled sand. Twenty-four 14-cm^3 packets of the sand (47 μCi/cm^3) were placed in a 130-m-long line, perpendicular to the beach, and a survey was made 23 hr later. One hour after that, thirty-six 14-cm^3 packets (22 μCi/cm^3) were placed in a 175-m-long line, and a survey was again made a day later. The data obtained in this test are considered reliable. Some seaward movement of sand was indicated, strong downcoast (lateral) and slight upcoast movement as well as zonal differences in movement rate were evident. A similar test in February 1969 was terminated because of adverse weather conditions.

COMPARISON OF RIST WITH OTHER TRACING METHODS*

According to the literature, sediment paths have been followed with various stable tracers, e.g., crushed glass or brick or natural sediment coated with bright paint or a fluorescent dye. Also, various radioisotopes have been used—formed in situ by irradiating the natural sand or by attaching a radioisotope to the sand in some way. The advantages of the radioisotopic method include the possibility of detection of the tracer in situ even when partially buried and the relative simplicity of the identification procedure once the detector system has been perfected.

Although ^{198}Au has been used before, especially in determining dispersal pattern of silts and clays, the gold-labeling technique developed for the RIST project is a considerable improvement over old methods since the radionuclide fixed to the sand grain and detection of the isotope definitely show the location of a labeled sediment particle. The ^{133}Xe-labeling technique was developed specifically for the RIST program and is new to the sediment-tracing field.

In earlier studies, a sled device was used to carry the radiation detector—Geiger–Mueller tubes or scintillation crystals—over the bottom of the water body. Hundreds of pounds of labeled sand were often used, and considerable time was required for the surveys. By comparison, with the radioisotopic technique used here an area 900 by 1300 ft was surveyed in 44 min. The survey started 2 hr after injection of the isotope and extended from 3 ft inshore of the water line to 15 ft beyond it.

Thirteen hundred data points, collected at 2-sec intervals, were analyzed, and about 20 thousand ft^2 was actually sampled. In another test, 3300 by 1600 ft was surveyed in less than 2 hr—126 hr after injection of the isotope. The survey extended from 6 ft on the beach to 15 ft seaward of the surf zone, and 3 thousand data points were collected over a track nearly 20 thousand ft long.

Further, a literature search did not locate other studies in which the beach face, inshore zone, and the offshore bottom were synoptically examined, others being confined to a beach face or deep water. The earlier inability to look at the inshore zone, the zone of maximum transport, has been due to the use of vehicles and detector systems that were not rugged enough to withstand the heavy forces of the surf zone. The rotating-cylinder detector system[1] developed for RIST solves this problem. The four-detector array in this system provides a high degree of sensitivity, increases detection capability, and examines a larger area and thus improves the counting statistics. The RIST data-acquisition system, which automatically collects and correlates time, position, and radiation counts, is unique in the field of sediment tracing.

*See also "Comparison of Radioisotopes and Fluorescent Dyes for Estuary Pollution-Control Studies" by H. P. Raaen in this issue of *Isotopes and Radiation Technology*.

REFERENCES

1. F. N. Case, E. H. Acree, and H. R. Brashear, Detection System for Tracing Radionuclide-Labeled Sediment in the Marine Environment, this issue of *Isotop. Radiat. Technol.*
2. N. H. Stephens, E. H. Acree, and F. N. Case, Evaluation of Four Methods Using ^{198}Au for Surface Labeling and a New Technique for Simulating Mass Labeling, USAEC Report ORNL-4338, Oak Ridge National Laboratory, December 1968.
3. E. H. Acree and F. N. Case, Method for Tagging Sand with a Gaseous Radioactive Isotope, U. S. Patent No. 3,426,205, Feb. 4, 1969 (filed Sept. 28, 1967).

4. E. H. Acree, F. N. Case, N. H. Cutshall, and H. R. Brashear, Radioisotopic Sand Tracer Study (RIST) Status Report for May 1966–April 1968, USAEC Report ORNL-4341, Oak Ridge National Laboratory, 1969 (included in Ref. 5*b*).
5. *a*. D. B. Duane, Tracing Sand Movement in the Littoral Zone: Progress in the Radioisotopic Sand Tracer (RIST) Study, July 1968–February 1969, Miscellaneous Paper No. 4-70, U. S. Army, Corps of Engineers, Coastal Engineering Research Center, August 1970.
b. D. B. Duane and C. W. Judge, Radioisotopic Sand Tracer Study, Point Conception, Calif., July 1966–June 1968, Miscellaneous Paper No. 2-69, U. S. Army Corps of Engineers, Coastal Engineering Research Center, May 1969.

Radioisotopic Gages in Pollution Control

By Martha Gerrard

Supplementary Keywords: *pollution; sedimentology; measurement and control system; hydrology; water and waste treatment;* ^{60}Co; ^{106}Ru.

Abstract: *Two Division of Isotopes Development–developed gages that can be used in pollution studies are described, and several developed elsewhere are mentioned.*

Gages in which attenuation or backscattering of radioisotopic radiation is used to measure thickness, level, density, and other properties of product streams and products have been widely applied in industry. Two such gages developed in work done under contract to the USAEC Division of Isotopes Development which can be used in pollution work are the eggshell-strength gage and the suspended-sediment gage described in earlier issues of *Isotopes and Radiation Technology*. These gages are summarized here. Instruments based on ^{85}Kr for determining automobile-exhaust pollution, a mercury-substitution gage for determining SO_2 in stack gases, and a beta-absorption monitor for atmospheric particulates are described in detail elsewhere in this issue of *Isotopes and Radiation Technology*.

EGGSHELL-STRENGTH GAGE

The eggshell-strength gage,[1] developed by R. S. Pressly of Oak Ridge National Laboratory and P. E. James of the Agricultural Engineering Research Division, U. S. Department of Agriculture, Beltsville, Md., can be used to study the effect of pesticides on bird populations. It is based on the principle that the amount of beta radiation backscattered from an intact eggshell is a function of the shell strength.

Ecological studies have shown a correlation between the decline in population of certain raptorial birds and their exposure to pesticides, possibly due to the thinner shells of the eggs of the exposed birds. Although this gage (Fig. 1) was not designed to measure thickness per se, if the readings on the gage should correlate with thickness, it would be useful in pesticide studies. Only 1 to 2 min is required to make a measurement with the gage, and the test is nondestructive. The gage was laboratory tested[2] to determine the effect of base material—shell membrane, albumen, and yolk—on the amount of radiation backscattered by the shell, and the gage readings were found to correlate significantly with shell thickness (Fig. 2).

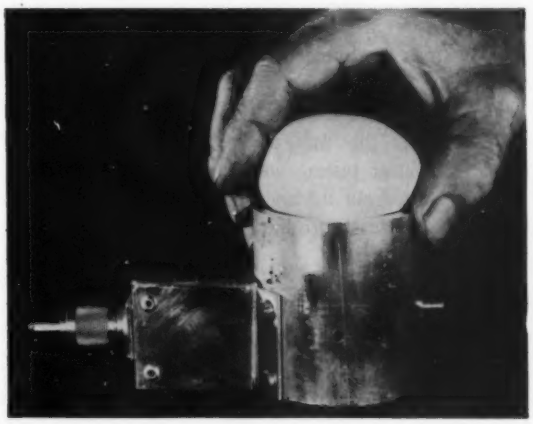

Fig. 1 Beta-backscatter gage for determining eggshell strength. The gage contains 10 μCi of ^{106}Ru, whose beta radiation is directed toward the eggshell, and the amount of radiation reflected (i.e., backscattered) from the shell is measured.

The laboratory data have now been tentatively confirmed in a field test[3] with Japanese quail fed

XUM

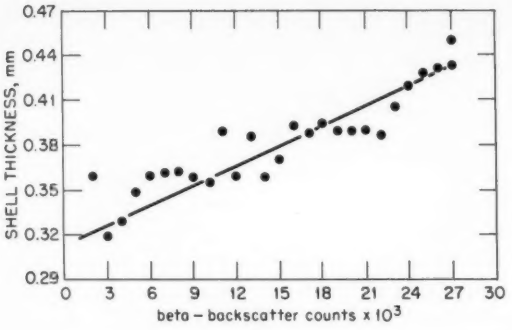

Fig. 2 Relation between shell thickness and beta-backscatter readings for dried shells.

DDT.* The thickness of the shells of their eggs, as measured with a micrometer, was less than in normal eggs, and the calcium content determined by absorptiometry was lower. Beta-backscatter measurements of strength were then made and found to correlate well with both thickness measurements and calcium-content determinations. The nondestructive, beta-backscatter method is described as being "rapid. . . and extremely suitable for experiments of this type."

Similar tests were made[4] on the shells of eggs of Mallard ducks fed DDE (a stable metabolite of DDT) and DDD. In this case the only shells available for beta-backscatter testing were those saved for residue analysis, but again thickness measurements correlated well with backscatter counts.

SUSPENDED-SEDIMENT GAGE

A self-referencing gage (Fig. 3) based on ^{109}Cd for measuring the amount of suspended solids in a flowing stream was developed[5] by Bach Sellers of Panametrics, Inc., and J. R. McHenry of the U. S. Department of Agriculture, Agricultural Research Service, Soil and Water Conservation Research Division, Oxford, Miss. The gage operates by alternately measuring the transmissions of X radiation through river water and through distilled water and then comparing beam intensities. If no appreciable amount of dissolved material is present, the ratio of the two values is a function of the suspended-sediment concentration. Since a ratio rather than an absolute value is recorded, errors due to source decay and to changes in the X-ray detector response, the electronics, and the liquid density are eliminated. Measurements are made continually.

In field tests[6] in flood flows of up to 5000 ft^3/sec with sediment concentrations as high as 14,000 ppM, suspended sediment-concentration measurements were comparable to those obtained by conventional (sampling) techniques.

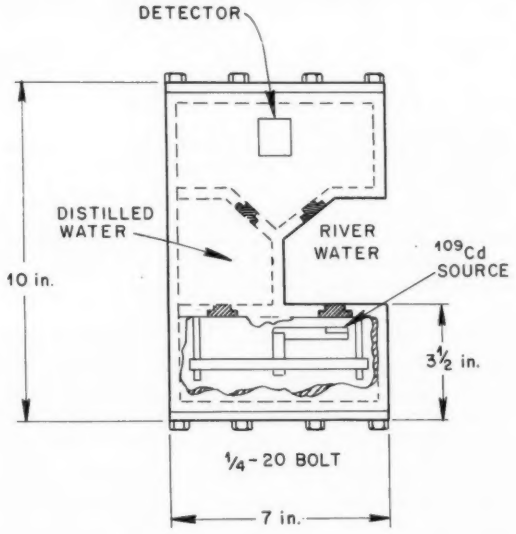

Fig. 3 Measuring head of self-referencing, single-probe, suspended-sediment–concentration gage.

OTHER GAGES

Of the many gages that could be used in pollution control, only a few are mentioned here as examples. The literature is sparse, but undoubtedly a number of gages used in sewage and pollution work have simply never been described.

The density of various streams in sewage-treatment plants should be known for most efficient operation and transfer of material from one stage to another. A gage has been patented[7] which controls the density of sludge transferred from one stage to another. It contains a source of penetrating radiation (gamma, X, beta, or neutron). The density of the sludge is indicated by readings on a radiation counter on the opposite side of the stream. When the density differs from a preset value, an alarm system is activated. A

*DDT is 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl)ethane: Cl–C$_6$H$_4$–CHCl$_3$–C$_6$H$_4$–Cl. DDE is 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethylene. DDD is 1,1-dichloro-2,2-bis(*p*-chlorophenyl)ethane.

XUM

Polish development[8] is a gage containing 20 mCi of ^{60}Co and a NaI(Tl) scintillation counter that detects the gamma radiation transmitted by an effluent stream and indicates when such effluent thickens.

A radioisotopic gage for determining sludge levels in a sewage plant has been described in Czechoslovakia.[9] Operation of the gage is based on attenuation of radiation by the sludge in the tank. The original article gives a detailed design of the instrument and instructions for its use.

REFERENCES*

1. Eggshell-Strength Determination by a Beta-Backscatter Technique, *Isotop. Radiat. Technol.*, 5(4): 284-286 (1968).
2. *a.* P. W. Voisey and P. E. James, Factors Affecting the Performance of the Beta Backscatter Eggshell Measurement, *Can. Agr. Eng.*, 12(1): 48-51 (1970).
   b. J. Bitman, H. C. Cecil, S. J. Harris, and P. E. James, Relationship of Beta Backscatter Counts to Eggshell Thickness and Calcium Content, *Poultry Sci.*, 48: 2184-2185 (1969).
3. J. Bitman, H. C. Cecil, S. J. Harris, and G. F. Fries, DDT Induces a Decrease in Eggshell Calcium, *Nature*, 224: 44-46 (1969).
4. R. G. Heath, J. W. Spann, and J. F. Kreitzer, Marked DDE Impairment of Mallard Reproduction in Controlled Studies, *Nature*, 224: 47-48 (1969).
5. Martha Gerrard, Radioisotope Gage for Determining Suspended-Sediment Concentration in Streams, *Isotop. Radiat. Technol.*, 4(4): 358-362 (1967).
6. *a.* C. E. Murphree, G. C. Bolton, J. R. McHenry, and D. A. Parsons, Field Test of an X-Ray Sediment-Concentration Gage, *Amer. Soc. Civil. Eng. Proc., J. Hydraul. Div.*, 94(Hy2): 515-528 (1968).
   b. Ibid., 96(Hy1): 515-516 (1970).
7. C. O. Badgett (to Industrial Nucleonics Corporation), Radiation Density Gage Control of Sludge Transfer Operations in Sewage Works, U. S. Patent No. 3,128,786, Apr. 14, 1964 (filed Nov. 27, 1959).
8. R. Burck, R. Lach, J. Lash, and J. Sawicki, Isotope Gage for Thickening of Effluent Waters (in Polish), *Przegl. Gorniczy*, 13(3): 185-189 (1962).
9. J. Szucs, Use of Radioisotopes for Determination of the Sludge Level (Slovak), *Vod. Hospod.*, 19(3): 82-83 (1969) (*Chem. Abs.* 72: 70395*t*).

*Some of the references used in preparing this article were supplied by P. E. James and J. R. McHenry.

Measurement of Sulfur Dioxide in Stack Gases: Design, Fabrication, and Test of Demonstration Instrument†

Summarized by R. H. Lafferty, Jr., P. S. Baker, and Martha Gerrard

Supplementary Keywords: *industry, utility; industry, primary metal; pollution; atmosphere; chemistry, analytical; absorptiometry; instrument;* ^{109}Cd.

Abstract: *The technical and economic feasibility of continuously measuring the sulfur dioxide content of stack gases comprising a technique that involves complexing part of the mercury in* $HgCl_2$ *with the* SO_2 *(as sulfate) followed by absorptiometric measurement of the mercury using a radioisotopic (*^{109}Cd*) X-ray source was established. In extensive laboratory and field tests, a working model of the proposed instrument measured sulfur dioxide from 100 to 4000 ppM with an accuracy of 2% and a response time of about 15 min. The major advantages of the instrument are simplicity of sampling and ease of maintenance. Several existing nonnuclear techniques are described briefly for comparison.*

Despite the predicted future impact of nuclear power, fossil-fuel use will undoubtedly increase until at least 2000 A.D. National concern with air pollution will force users of fossil fuels to control and monitor their emission of stack gas into the atmosphere, but no SO_2 monitor now available operates continuously and reliably (see Appendix), with low maintenance, directly at the stack. Dispersive and nondispersive ultraviolet-absorption gages are being field tested, but some engineering difficulties still must be resolved. The flame-spectrophotometric technique is in an early stage of development and testing, and the remote-sensing devices that may be used for spot checking by carefully aiming the sensor at a portion of the plume are

†Summary of USAEC Report NYO-3882-2, May 28, 1970, by Robert J. Pfeifer, Industrial Nucleonics Corporation, 650 Ackerman Road, Columbus, Ohio 43202. Drawings of the instrument are available as USAEC package CAPE-2003.

unsuitable for continuous measurements. Nondispersive infrared gages have been evaluated without conclusive results, and chemical systems (e.g., electrolytic) do not appear to have been used.

Following discussions with coal and power representatives in 1967, the AEC Division of Isotopes Development started to investigate the need for a system to routinely monitor the amount of sulfur passing through an industrial stack. Preliminary studies were made of existing methods of measuring sulfur in stack gases and of the feasibility of a mercury-complexing technique with a radioisotopic gage for measuring the amount of substituted mercury. Based on the results of these studies, DID contracted with Industrial Nucleonics Corporation to develop and test a working model of a mercury-complexing system with a radioisotopic gage for measuring SO_2 in stack gases. The design, fabrication, and testing of a prototype instrument based on this system are described here.

SYSTEM DESCRIPTION

The system was designed to automatically and continuously monitor the SO_2 in a side stream from the stack. The sulfur (low atomic number), as SO_2, complexes with part of the mercury (high atomic number) in Hg_2Cl_2, forming water-soluble *bis*-sulfitomercurate $[Hg(SO_3)_2]^{2-}$:

$$Hg_2Cl_2 + 2H_2O + 2SO_2 \rightarrow Hg(SO_3)_2^{2} + 2Cl^- + 4H^+ + Hg \text{ metal}$$

A sodium acetate–acetic acid buffer in the reaction-cell medium maintains the *p*H at about 5, which is necessary for the reaction to proceed satisfactorily.

The water solution of the mercury complex flows through a filter to a measurement cell, where it intercepts the 22-keV X-ray beam from a 50-μCi ^{109}Cd source. The amount of X radiation absorbed, as shown by a counter on the other side of the mercury solution, is quantitatively related to the amount of Hg(II) in the solution.

A transmission method rather than an X-ray-fluorescence method was selected for measuring the released Hg(II) because, at the lowest mercury concentration that might occur, the count rate must be determined with a precision of ±0.1%. In preliminary tests, this precision was achieved with the transmission system, but interference from scattered X rays in the X-ray-fluorescence method resulted in an error that was an order of magnitude greater.

INSTRUMENT CONSTRUCTION AND OPERATION

A mercury-complex system, incorporating a radioisotopic radiation detector (Fig. 1), was fabricated in accord with the results of stoichiometry tests and preliminary design studies. The working model, 28 in. high by 18 in. wide by 15 in. deep, weighs approximately 90 lb. The interior is divided into two compartments—an ambient-temperature compartment where the pump motors and the data-processing system are located and a constant-temperature compartment where the entire reaction and measuring systems are located. The temperature of this compartment is maintained at 115 ± 2°F by a fan–heater combination, which circulates air within the instrument.

Reaction Section

The glass reaction cell, which is 20 in. high and approximately 3 in. wide and deep, has two sections, connected by a ground-glass joint. In the lower section are the reaction chamber itself, the input port for stack gas, and the output port for the $[Hg(SO_3)_2]^{2-}$ stream. In the upper section are an expansion chamber to take care of possible frothing of liquids and the input ports for water and acetate buffer.

Metering pumps for water and acetate buffer and a vacuum pump through which the stack gas is drawn are shown on the right in Fig. 1. A 1-liter reservoir for the buffer, at the top, must be filled about once a week to accommodate a flow of 0.1 ml/min. A water-holding tank, not shown in Fig. 1, reduces the pressure to the water-metering pump from line pressure to less than 20 in. H_2O. The gas flow to the reaction cell is determined by a network of orifices and solenoids.

Measurement Section

The measurement system, located at the bottom of the instrument, consists of a radiation source, a measurement cell, and two scintillation-multiplier phototube detectors (Fig. 2). The fluid containing the $[Hg(SO_3)_2]^{2-}$ complex enters the measurement cell from the top.

The 50-μCi ^{109}Cd X-ray source is located in a 0.5-in.-diameter by 0.25-in.-deep capsule. The source material is electrodeposited on a nickel substrate and covered with a thin electrodeposited nickel layer, and the wafer is encapsulated in aluminum. The detectors—Harshaw integral line type, each consisting of a NaI scintillator, 1.32 in. thick by 1 in. in diameter, joined to an Amperex XP1010 multiplier photo-

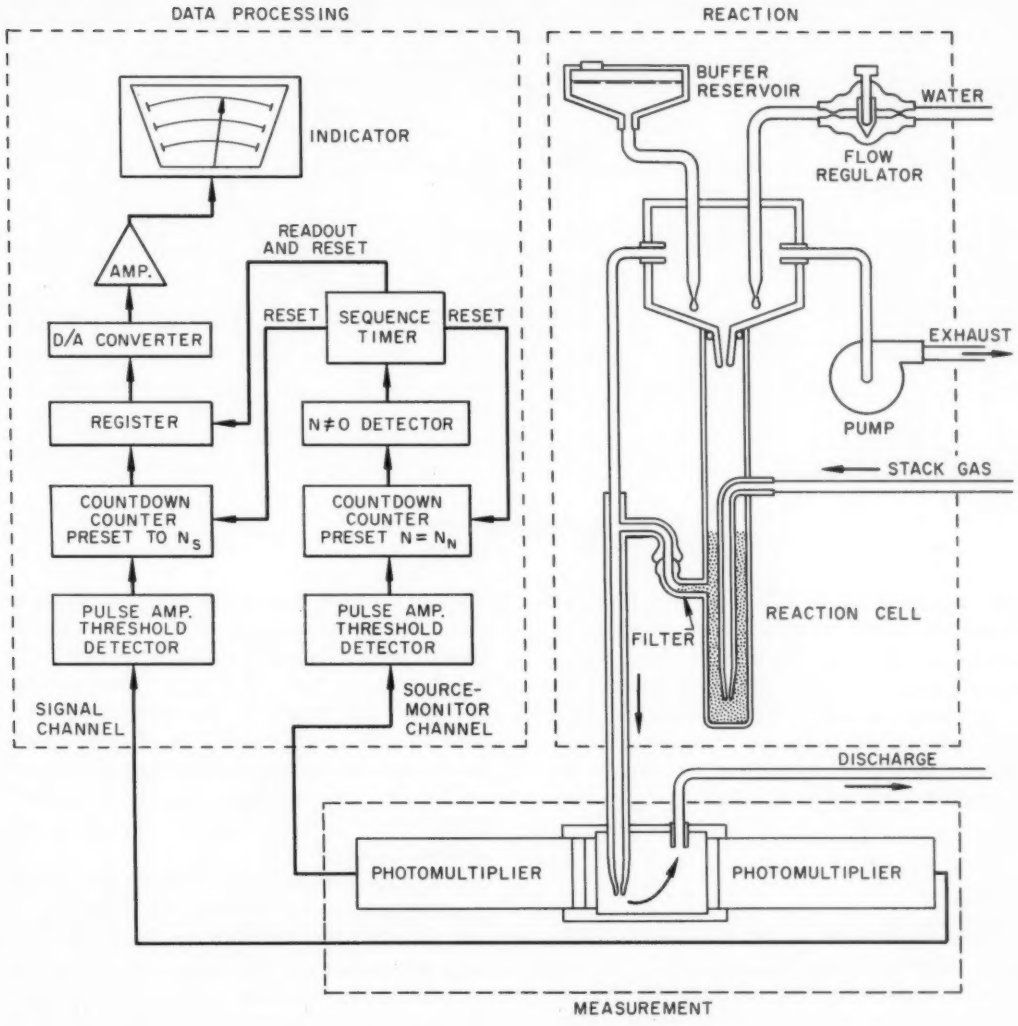

Fig. 1 The SO_2 detector.

tube—are mounted directly on the measurement-cell structure. The entire measurement system is located in the thermally controlled chamber.

Data-Processing System

The data-processing system is shown on the left in Fig. 1. The solid-state data-processing electronics are contained on 15 printed-circuit cards, on two pre-amplifier boards, and in several power-supply packages. The data-processing electronics operate in a digital mode until the final conversion to analog form, which is supplied to the output meter and recorder. Interferences from r-f and internal digital-pulse sources were eliminated by the use of shields.

Gage Operation

For operation, the gas flow rate through the system is selected by a range switch on the instrument panel. The low range corresponds to 0.15 liter/min, the medium to 0.5 liter/min, and the high to 1.5 liters/min. Beneath the range switch is a needle-valve control through which the flow rate can be adjusted further. A

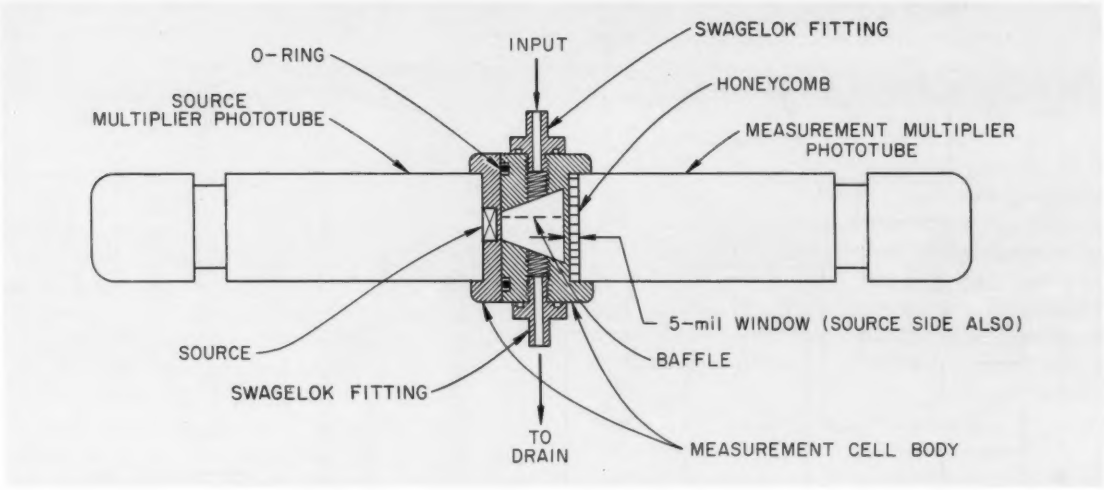

Fig. 2 Measurement system.

Plexiglas inspection port is located between the range switch and the needle valve, and directly above the range switch is a meter that indicates the SO_2 content of the stack gas. Directly below the needle valve are output taps for use in monitoring various signals within the instrument, including the SO_2-measurement output. Various electronic controls are provided on the panel, together with switches to control gage conditions.

TESTING

The instrument was tested extensively in the laboratory, throughout the full range of SO_2 concentrations to be expected in field application, and on an experimental furnace. The furnace used either pulverized coal or gas as fuel, with typical levels of fly ash, nitrogen oxides, etc., in the off-gas. The SO_2 content of this gas was varied by burning coals of different sulfur contents and by introducing SO_2 into the air intake for either coal or gas fires. The furnace tests culminated in a continuous 1-month test. Sensitivities and response times were determined for different SO_2 concentrations and for different gas and liquid flow rates.

Typical responses of the instrument to step changes in SO_2 concentration on medium- and low-sensitivity scales are shown in Fig. 3. Since gage response was characterized by extremely good sensitivity, data were collected for medium and low sensitivities only, i.e., 20 to 50 ppM SO_2 per division and greater than 50 ppM SO_2 per division, respectively. Occasionally an "overshoot" response occurred when the instrument was turned on from the standby state or immediately after introduction of fresh Hg_2Cl_2 into the reaction cell. The exact nature of the phenomenon is not fully understood, but it is thought to result from a mixing of fluid in the reaction cell after that fluid had settled and stratified during a significant period of quiescence. The condition did not occur often enough to warrant systematic study.

Typically, the response time of the instrument (time for the output to reach 65% of its final value) was about 15 min for both laboratory and furnace tests; this included a time lag of about 5 min and an exponential time constant of about 10 min. The lag time is an inverse function of both the gas and liquid flow rates, decreasing as these rates increase. For gas flow rates of about 0.15 liter/min, the time lag was long because of the time required to draw the sample into the instrument. This component of the lag time could be eliminated by a sampling system in which a large gas volume is drawn past the instrument, a small amount of the gas being drawn through the instrument. The exponential time constant is a function of the fluid flow rate; rates of 2.0 ml/min resulted in time constants of 8 min and rates of less than 1.0 ml/min in constants of less than 15 min.

Throughout all the testing, maintenance was concerned primarily with the reaction system. Unfortunately, miniaturization of the flow system, done to decrease the response time, degraded the reliability of

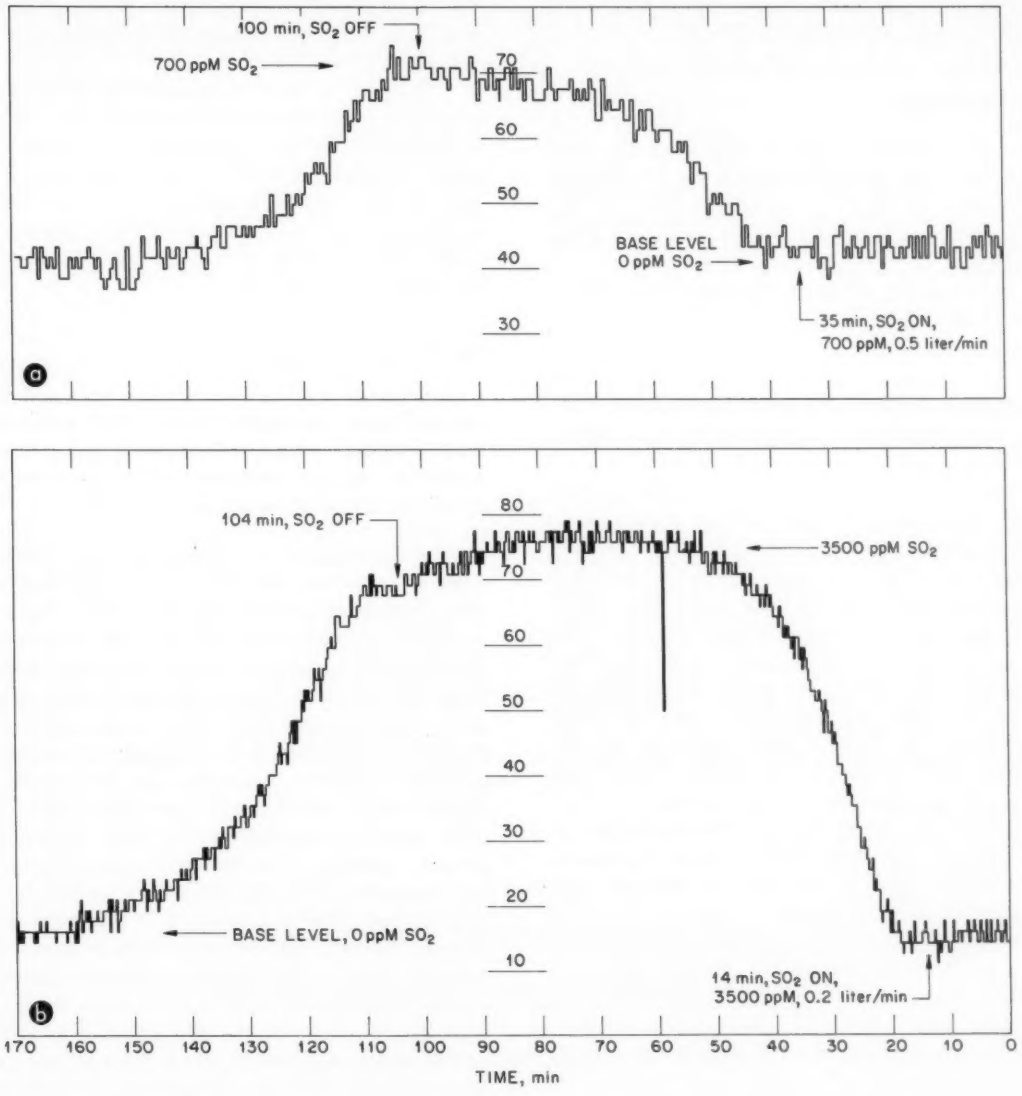

Fig. 3 Tests of (*a*) Aug. 11, medium sensitivity, 24 ppM SO_2 per division; and (*b*) Aug. 26, low sensitivity, 59 ppM SO_2 per division.

the chemical system. Clogging of the glass frit in the reaction cell necessitated frequent shutdowns, and occasionally excessive frothing carried Hg_2Cl_2 into the expansion chamber, where it did not satisfactorily react with the SO_2. Bubbles occasionally were trapped in the measurement cell. Nevertheless, the goal of 7 days of trouble-free operation was achieved several times during the furnace tests.

CONCLUSIONS

It appears that this system (1) will not require cleaning or dewatering of the stack-gas sample; (2) is insensitive to fly ash; and (3) will eliminate problems of relative humidity that hinder other SO_2-measuring systems. The method will be wholly continuous, the chemical yield will be more than 90%, and the

XUM

measurement range will be 100 to 4000 ppM, which covers the range of 0.1 to 5% sulfur in coal.

REFERENCES

1. The three feasibility studies are all reported in a single document, Studies of the Measurement of Sulfur Dioxide in Stack Gases, USAEC Report TID-25137, Industrial Nucleonics Corporation, Nuclear Science Division of International Chemical and Nuclear Corporation, and Texas Nuclear Corporation, Jan. 29, 1968.
2. G. Kotnick and H. F. Sheck, Monitoring SO_2 in Stacks, *Instrum. Technol.*, 16(9): 52-55 (September 1969).

APPENDIX: NONNUCLEAR METHODS FOR SULFUR DIOXIDE DETERMINATION

Comparative features of various nonnucleonic gages for SO_2 measurement in stack gases are discussed both in the feasibility-study reports[1] and in the literature. No significant progress seems to have been made since these evaluations. Users of these gages want instruments which meet user requirements—probably to be defined by air-pollution agencies—and which are reasonable in cost. The performance specifications for gages are available from the manufacturers, but they really amount to manufacturers' claims and are not complete until all engineering problems have been solved and the gage has been field-tested for a long enough time to solve all the applications problems. A true evaluation should be based on users' experience, and relatively little in the way of unbiased user evaluation is currently available.

Nonnucleonic techniques that have been used for stack-gas SO_2 measurements are chemical and spectrometric. Except for some isolated examples, no significant effort has been expended on continuous on-line measurements based on chemical analysis.

Chemical Methods

The only example of a chemical continuous-monitoring measurement system is a bromine–bromide electrolytic device developed in the Netherlands. In this device, in which bromine is continuously reduced to bromide by SO_2 in an electrolytic cell, a feedback system maintains the bromine level constant; the current supplied to the cell is therefore a function of the rate of bromine reduction by SO_2 and hence is proportional to the SO_2 content. Critical components include a particulate filter and a catalytic filter for removal of interfering gases. Extensive field evaluation of the instrument will be needed to determine whether these components are satisfactory for the precision and maintenance required.

The gage has been extensively field-tested and is being used for determining low concentrations of SO_2 in ambient air, but its application to stack-gas analysis, where concentrations of interfering particulates and gases are relatively high, has not been demonstrated. Significantly, the gage is being tested in the Netherlands within a system of computer-coordinated SO_2-measurement instruments.

Spectrometric Methods

A number of spectrometric techniques have gone through both analytical evaluation and preliminary testing. Several versions of infrared gages have been carried through the complete cycle of evaluation, and some ultraviolet gages are being field-tested.

Nondispersive Infrared-Absorption Gage (Beckman, Leeds and Northrup, and MSA). These gages have been most extensively tested. At least three types are available:[2] (1) chopper-stabilized dual source–dual cell–differential detector system; (2) single source–single cell–negative filter–sensitized detector system; and (3) chopper-stabilized dual source–dual cell–positive/negative filter–differential detector system.

Various filtering techniques and the features of common-mode rejection built into these systems partially eliminate interferences by water vapor, CO_2, nitrogen oxides, and dust. However, these interferences and instrument-drift problems, when applied to the measurement of trace quantities of SO_2, are severe enough that some interfering constituents have to be removed by a rather elaborate sample-conditioning system. When dust or water vapor is to be eliminated from the sample before the measurements, some SO_2 may be simultaneously removed in the sampling train. A nondispersive infrared gage may find its application in continuous, on-line measurements of minor constituents of stack gas or in laboratory analysis of trace compounds but probably not in continuous, on-line measurements of trace quantities of SO_2 in stack gas.

Nondispersive Ultraviolet-Absorption Gage (Du Pont). Water vapor and CO_2 do not interfere with the SO_2 measurement in the ultraviolet region of the spectrum. Interference by the trace quantities of nitrogen oxides is eliminated by the dual-wavelength approach. Interference by dust is partially eliminated by using a single source–single cell–dual wavelength technique. However, because of the broadness of the

XUM

absorption bands in the ultraviolet region of the spectrum, the difference in wavelengths between the reference and measuring channels has to be large, and therefore the attenuation of radiation due to scattering by dust does not completely cancel out. Any significant dust interference may be eliminated by filtering the gas at an elevated temperature (to prevent water condensation and simultaneous partial removal of SO_2). Some engineering problems still remain to be solved. For example, many ultraviolet sources have a rather short service life and questionable spectral stability. Furthermore, the use of two independent detectors and amplifiers for the two separate channels makes the stability of the system rather poor.

Dispersive Ultraviolet-Absorption Gage (Barringer Research). In principle, this system, which is inserted directly into the stack-gas stream, offers advantages over nondispersive techniques with respect to the elimination of interference by nitrogen oxides and dust. The elimination of dust interference is probably not complete.

This system has engineering problems similar to those of a nondispersive system with regard to obtaining a source of sufficient intensity and long service life, and the sensitivity is probably not so high as for the nondispersive system. The stability problems of detectors and amplifiers in the nondispersive system are replaced by manufacturing problems associated with precision mechanical parts and the correlation mask (i.e., a photographic plate of an SO_2 spectrum).

Ultraviolet-Emission Gage (Melpar). A highly sensitive system, utilizing a hydrogen-flame photometer in which the stack gas is burned, provides some advantages over the ultraviolet-absorption techniques since emission at wavelengths corresponding to SO_2 is quite intense. The hot stack gas must be mixed with hydrogen and oxygen at the sampling point, and the dew point of the sampled gas must be maintained above ambient temperature so that moisture does not condense in the sampling tube. Particulate matter must be removed from the gas to eliminate interfering emission wavelengths. In general, interference from other gases is the same as for the nondispersive ultraviolet-absorption technique, although the existence of additional emission wavelengths provides a greater choice for the instrument.

Remote-Sensing Devices. At least two remote-sensing devices have been proposed. One instrument produces an infrared-emission spectrum and correlates it with the amount of SO_2 in stack gas by aiming a sensor at the stack plume. The other instrument (Barringer) uses the correlation-mask technique with sunlight as the radiation source. Accurate quantitative measurement may, however, be difficult owing to the changing atmospheric conditions and plume configuration.

Kryptonate-Based Instrument for Detecting Automobile-Exhaust Pollutants*

Summarized by Martha Gerrard and R. H. Lafferty, Jr.

Supplementary Keywords: *tracer; radiorelease; industry, transportation; pollution; atmosphere; ^{85}Kr.*

Abstract: *The technical feasibility of using ^{85}Kr to detect pollutants—carbon monoxide, nitrogen oxides, and hydrocarbons—in automobile exhausts in concentrations below the projected 1975 standards was demonstrated. In a laboratory test system, 13 ppM C_3H_8, 2 ppM NO_X, and 125 ppM CO were detected by a PtO_2 Kryptonate sensor, a hydroquinone clathrate sensor, and a $PdCl_2$ Kryptonate sensor, respectively. Several nonnuclear methods for detecting exhaust pollutants are described briefly for comparison.*

Air pollution, a worldwide problem today, is due in large part to automobile exhaust. Control of the release of this exhaust, which has not been very stringent in the past, is expected to increase drastically by 1975. Then the maximum permissible concentrations of hydrocarbons, nitrogen oxides, and carbon monox-

*Summary of Kryptonate-Based Instrumentation Development for Automobile Exhaust Pollutants, USAEC Report NYO-4069-1, Philip Goodman and Thomas Donaghue, Panametrics, Inc., 221 Crescent St., Waltham, Mass. 02154.

XUM

ide—the chief components of the exhaust emission—are expected to be 50, 250, and 5000 ppM, respectively. An inexpensive portable instrument for determining these pollutants is needed for use, for example, by highway patrols. However, a survey of the literature from manufacturers of exhaust-testing instruments (see Appendix) indicated that the only one on the market which can detect all three pollutants is nonportable, and some of the ones used for only one or two components are not very reliable and cost up to $3000.

Obviously, an inexpensive and portable instrument that could rapidly detect all three of these pollutants is highly desirable. Panametrics, Inc., under contract to the AEC Division of Isotopes Development, undertook to develop such an instrument, based on the radiorelease technique using a ^{85}Kr-containing reagent. Radiorelease techniques based on the use of ^{85}Kr clathrates or Kryptonates* have been developed for determining a number of substances, including several gases. This technique therefore seemed promising for the automobile-exhaust problem. Its two chief advantages are: (1) the ^{85}Kr is contained only in the surface layers of the reagent, which gives it high sensitivity to trace gas concentrations, and (2) the ^{85}Kr can be introduced into a great variety of solids, which makes it possible to minimize interference effects.

The development of an instrument system and tests on it which showed the technical feasibility for detecting each of the three chief pollutants in automobile exhaust are described here.

DETECTION SYSTEM

The radiorelease technique is one in which a radioactive element is released from a radioactive reagent by the substance under study. The amount of radioisotope released is quantitatively dependent on the amount of test substance present. In the automobile-exhaust-pollutant detection system developed in this project (Fig. 1), there were three sensors containing ^{85}Kr-labeled reagents—one with PtO_2 Kryptonate for detecting hydrocarbons, one with hydroquinone clathrate for detecting nitrogen oxides, and one with $PdCl_2$ Kryptonate for detecting carbon monoxide. The test gases were fed into the system from stock supplies after being diluted to suitable concentrations for testing. Propane was available as a gas, but the higher hydrocarbons studied, which are liquids at room temperature, were obtained in the form needed for use by passing air slowly through liquid batches of these substances until it was saturated.

A Geiger–Mueller counter downstream from the sensors showed the amount of activity in the effluent stream, i.e., the amount of ^{85}Kr released in the sensor chamber by the pollutant under study. In the system described here, if fully developed, the three pollutants would be detected sequentially, the G–M chamber being flushed after each measurement. If simultaneous detection should be desired, a counter would have to be provided for each sensor, which would increase the cost.

TESTS

Hydrocarbon Detection

The Kryptonate of PtO_2 had been shown[2] to be suitable for detecting hydrocarbons and therefore was selected for the hydrocarbon sensor in the automobile-exhaust instrument. The reagent, which was prepared by the method given in Ref. 2, was placed in the sensor cell, where an atmosphere of about 2% O_2 in N_2 was maintained for most runs. The absence of oxygen increased the system sensitivity.

With hexane as the hydrocarbon at a concentration of 1000 ppM and a constant flow rate of 800 cm^3/min, the sensor response varied linearly with temperature from 350 to 550°F. The sensitivity at 500°F was sufficient for detection of 20 ppM C_6H_{14}, and subsequent experiments were done at this temperature. However, the system was more sensitive to propane at a higher temperature, which should be considered in the final design of such an instrument.

At 500°F, the sensor response varied linearly with a change in flow rate from 250 to 500 cm^3/min and leveled off at 800 cm^3/min. This last value was therefore used in subsequent runs. The sensor response also varied linearly with the hydrocarbon molecular weight from propane through octane within 10% error limits.

Reproducibility of the PtO_2 Kryptonate sensor was investigated by alternately passing a hexane stream and an air stream through the sensor cell at 2-min intervals for 26 complete cycles. The average signal recorded was 1987 counts/min, and the standard deviation was

*As considered here, both clathrates and Kryptonates are solids containing ^{85}Kr. A clathrate is made by crystallizing an organic compound from the molten state in the presence of a gas, which may be an inert one. Only ^{85}Kr in hydroquinone was used here. A Kryptonate (registered trademark of Panametrics, Inc.) is made by impelling krypton ions or pressure-diffusing krypton atoms into a solid, which may be any solid.

XUM

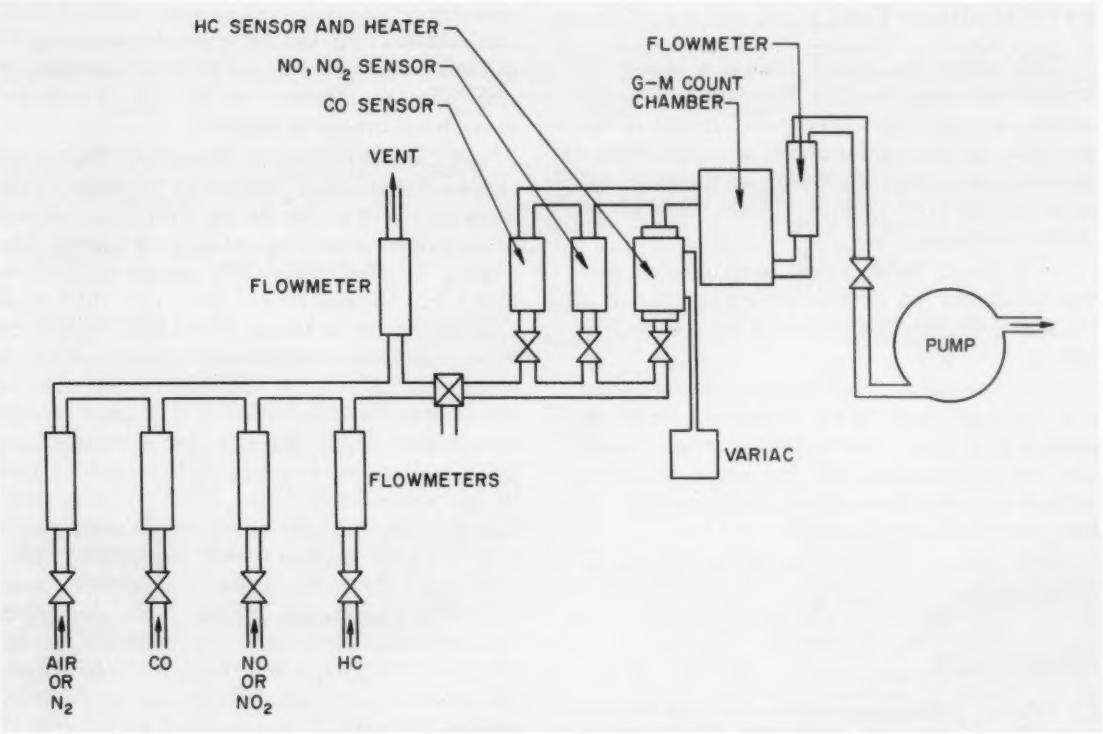

Fig. 1 Automobile-exhaust-pollutant test system.

8.6% of the average reading. For the particular ratemeter settings used, the error attributable to radioisotope counting statistics was about 7%, so that only about 1.5% of the observed variations are attributed to fluctuations in hexane concentration, sensor-response characteristics, and other experimental errors.

Nitrogen Oxides Detection

Several Kryptonates—including copper powder and osmium—were tested but were discarded when they were found to respond erratically to trace quantities of nitrogen dioxide at elevated temperatures. However, a hydroquinone–^{85}Kr clathrate responded with a constant signal and was adopted for the NO_x detector.

When nitrogen dioxide was passed through the sensor at room temperature at a flow rate of 800 cm^3/min, the sensor sensitivity was 200 counts/min per ppM of NO_2. With a flow rate of 200 cm^3/min, the sensitivity was 50 counts/min per ppM. Tests at rates up to 1600 cm^3/min showed a linear dependence of sensitivity on flow rate, indicating a 100% reaction efficiency with the clathrate in this range.

However, the clathrate sensor did not respond to NO. After a literature survey had yielded few candidates for a Kryptonate specific to NO, a means for converting NO to NO_2 was introduced into the system—a heated chamber inserted ahead of the sensor. The sensitivity to NO at a flow rate of 200 cm^3/min was found to be identical to that for NO_2, which indicates 100% conversion of NO to NO_2.

Carbon Monoxide Detection

The reagent selected for carbon monoxide detection was $PdCl_2$ Kryptonate. Nickel Kryptonate was tested but found completely unsuitable.

The $PdCl_2$ Kryptonate was sensitive to CO at room temperature at a flow rate of 100 cm^3/min, with a lower detection limit of about 125 ppM. This is a slow flow rate, but the 125-ppM value is about $\frac{1}{40}$ that required for the 1975 standards, and the 5000-ppM standard could be detected at a much faster flow rate and therefore with a response time of less than 10 sec.

SYSTEM ADVANTAGES

Each sensor was specific for the substance for which it was designed—PtO_2 Kryptonate for hydrocarbons, hydroquinone clathrate for nitrogen oxides, and $PdCl_2$ Kryptonate for carbon monoxide. When a gas stream containing one pollutant was passed through sensors designed for another pollutant, there was no significant response.

A laboratory simulation of combustion of commercial gasoline did not indicate any interference in the system by other components of the gasoline, e.g., lead.

The cost of the ^{85}Kr radiorelease system should be low. Although design is too preliminary for a cost estimate to be made, $350 to $500 per sensor channel does not seem unreasonable. The reagent should be available at a cost of $6 to $10 for 10 μCi, with 2.4 μCi being used per channel per week.

With ordinary care, there would be virtually no radiation hazard from the system.

REFERENCES

1. J. E. Carden, Radiorelease in Review with Special Emphasis on ^{85}Kr Clathrates and Kryptonates, USAEC Report ORNL-IIC-18, Oak Ridge National Laboratory, July 1969.
2. O. Cucchiara, P. Goodman, and R. Rex, Kryptonate Hydrogen Detection System, USAEC Report NYO-3735-1, Panametrics, Inc., May 1968.
3. R. K. Stevens and A. E. O'Keefe, Modern Aspects of Air Pollution Monitoring, *Anal. Chem.*, 42(2): 143A-148A (February 1970).

APPENDIX: NONNUCLEAR METHODS FOR AIR-POLLUTION MONITORING

Current instruments for air-pollution monitoring were adapted[3] from instruments in use in the chemical process industry in the mid-1950s and are chiefly based on measurement of color developed when a gaseous pollutant reacts with an aqueous solution of a color-forming reagent. Another type of instrument, using nondispersive infrared absorption for CO, suffers from serious water vapor and other interferences. Scrubbers have been introduced to obtain specificity, but they remove some of the pollutant of interest, are temperature dependent, and are short lived. Some sophisticated techniques that have been used include flame ionization detection and gas chromatography. These instruments require technically trained personnel for operation, are expensive, and are generally nonportable. A summary of pertinent information obtained from manufacturers' literature and personal contacts is given in Table 1. The table may not be complete because, in some cases, no literature was available or none was forwarded in response to requests.

Because the Ford Motor Company is making the Honeywell instrument available to its dealers, some detail on it is of interest. In this instrument a portion of the exhaust gases is burned on a hot filament. The resulting ultraviolet emission is representative of the total CO and hydrocarbon concentration, and a signal corresponding to an average hydrocarbon level in the exhaust gases is electronically subtracted from the total signal, resulting in a CO measurement. The response time is 20 sec for 95% of the reading. Alternatively, the exhaust gases can be passed through a charcoal filter before combustion to remove the hydrocarbons from the gas stream, so that the ultraviolet emission measures CO only. The hydrocarbon concentration scale is graduated only in bands labeled "acceptable," "marginal," and "excessive." The concentration range covered is not divulged, but the lowest measurable concentration can be estimated by considering that the CO scale is 0 to 5% and the accuracy is 5% of full scale. The minimum detectable concentration should therefore be 2500 ppM. If, as is assumed, the intensity of ultraviolet emission per unit concentration is the same for CO and for hydrocarbons, the minimum detectable concentration is higher by a factor of 50 than the projected 1975 standard for hydrocarbon emission.

Information and opinions from several installations active in the field of automobile-exhaust-pollutant measurement and control are summarized below.

U. S. Bureau of Mines, Bartlesville, Okla. This laboratory is principally concerned with detailed analysis of exhaust gases for hydrocarbon and hydrocarbon-derivative compositions under a variety of automobile-engine operating conditions. For instance, relative compositions of paraffin, olefin, and aromatic hydrocarbons; oxygenates (aldehydes, ketones, and acids); and acetylenes are determined as a function of engine operating speed, ambient temperature, and commercial gasoline types. The emphasis on experimental studies removes this organization somewhat from the operational requirements for monitoring automobile pollutants. For their own studies, they would like an instrument with a response time for hydrocarbons of 1 sec for transient conditions and 3 sec for average conditions. Specific suggestions for a Kryptonate test instrument were an easily replaceable filter in the sensor cell and a tamper-proof instrument

XUM

Table 1 Instruments on the Market for Determining Exhaust Pollutants in Air

Company	Gas	Conc.	Response time	Method	Compact or portable	Remarks
Dynasciences	NO_x	0–1000 ppM 0–5000 ppM	90% of reading in 15 sec	Electrochemical transducer	Yes	No response to HC, CO, CO_2, H_2O
Beckman/Scott	CO	0–10%		Infrared analysis	No	HC reported as *n*-hexane; built for automobile exhausts; ~6 by 4 ft
	HC	0–1000 ppM 0–10,000 ppM				
Honeywell	CO	0–5%	95% of reading in 20 sec	Ultraviolet emission	Yes	No amount of HC given, only marginal and/or poor combination; ~$700
	HC					
Technicon	CO	0–5 ppM 0–150 ppM		Wet-chemical analysis	No	Built in trailer
	NO	0–2 ppM				
	HC	0–5 ppM 0–25 ppM				
Devco Eng.	CO	40–500 ppM	30 sec	Catalyst and thermopile	Yes	
Bacharach Institute	CO	0–5%	95% of reading in 5 sec	Wheatstone bridge	Yes	Weight 38 lb; ~$400; HC interference
	HC	0–1000 ppM		Same	Yes	6 lb; HC as benzene; CO interference
Bendix	CO HC				No	Under development on contract from Post Office; to be set up in trailer; sensing techniques not chosen
Unico	CO	Down to 50 ppM		Color change with photo-optical detector	Yes	$300; sensor replacement after each CO exposure
Marquette Corp.	CO			Infrared	Yes	~$900
MSA	CO	0–10%	90% of final reading in 0.4 sec	NDIR*	Yes	~100 lb packed
	NO	0–2000 ppM				
	HC	0–1000 ppM 0–1%				
Autoscan, Inc.	HC			Catalytic combustion	Yes	Present cost ~$2000, projected ~$500; uses scrubber; being tested by Calif. Highway Patrol
Olsen	CO			NDIR	Yes	Cost ~$500; imported from Japan
Precision Scientific	NO_2	0–0.5 ppM 0–2 ppM	4 min for 63% of reading	Color formation in scrubbing solution		Laboratory use only; $1775
Beckman	NO^+ NO_2	0.3–3 ppM	10–15 min for 90% of reading	Color formation in scrubbing solution	Yes	29 lb
Beckman	HC	1 ppM to 2% full scale as CH_4	0.5 sec for 90% of reading	FID*	No	55 lb; ~$2000; H_2 generator required
Intertech	CO	0–10%	2 sec for 100%	NDIR	Yes	22 lb

*NDIR = nondispersive infrared; FID = flame ionization detection.

if it should be used in commercial garages. They believe that flame ionization detectors are usable in state inspection stations, are relatively easy to operate by trained personnel, and give reliable results but would be expensive to buy and operate.

California Air Resources Laboratory, Los Angeles. In California a law that all new cars must be inspected for exhaust emission at various motor speeds will be enforced in 1 to 2 years. For certified (i.e., equipped with emission-control devices when new) used cars, no universal inspection procedure exists other than visual inspection to make sure the device is connected. Fifteen test teams from the State Highway Patrol set up roadblocks in Southern California and randomly inspect cars. Each team needs an inspection instrument. There are also two voluntary test stations where engines are run through a seven-step cycle. It is desirable to be able to correlate this test with a two-speed test, perhaps at idle and 3000 rpm, the latter being important for emission of nitrogen oxides.

A quick test procedure is also needed for every car, whether tested on the assembly line or by a dealer. The desirable features of such an instrument are low cost, durability, dependability, 10 sec or less response time, and self-contained calibration. The California Air Resources Laboratory will cooperate in testing any instrument that is developed.

State of New Jersey. New Jersey is unique among the states in having already established state-run automobile inspection stations. There are 80 inspection lanes through one of which each car registered in the state must pass once a year for a safety inspection. Plans exist to equip these lanes with instruments to measure automobile pollutant levels. One such inspection instrument, designed by Scott Research Laboratories but not installed, consists of a dynamometer, a sampling system, a nondispersive infrared CO detector, flame ionization detector for hydrocarbons, and a computer-programmer to cause the automobile to run through an rpm-time cycle. New Jersey appears to be set on the infrared and flame ionization techniques for determining CO and hydrocarbons, respectively, despite the cost and complexity. The basic instruments cost about $3000 each, and addition of sampling systems brings the cost to about $10,000. An installation with dynamometer is estimated to cost at least $20,000. Response time is important since only about 1 min will be available to measure all three pollutants.

Suitable instruments for measuring NO_x are not available, and New Jersey is very much interested in the ^{85}Kr technique for this measurement.

Once inspection has been instituted, comparatively cheap instruments must be available for garages so that satisfactory repairs and adjustments can be made. The Kryptonate technique was thought to be very promising for this requirement. Licensing and handling of radioactivity, although not the forte of the inspectors, were not considered insurmountable. A portable instrument is needed for selected state-highway-patrol cars.

Production of Synthetic Fallout at Camp Parks

By William B. Lane*

Supplementary Keywords: *preparation, labeled material; meteorology; facility description; safety;* ^{90}Y; ^{140}Ba.

Editors' Note: The use of labeled sand and other particulate matter to study various environmental problems points up the need for suitable labeling techniques. The editors felt that the following article describes a procedure that might be suitable for a number of applications.

Abstract: *The preparation of sand labeled with* ^{140}Ba–^{140}La *and with* ^{90}Y *for use in synthetic-fallout studies is described together with the facility where the preparation is made.*

Technical data are needed to formulate survival measures for protection of the population in the event of a nuclear attack. Radioactive fallout is one such hazard, and experiments that can proceed under a nuclear-test moratorium require synthetic fallout.

Simulated fallout, whose properties simulate those of real fallout, has been prepared in the hot-cell facility at Camp Parks, Calif., for studies sponsored by the Office of Civil Defense at Stanford Research Institute.

*Engineering Systems Division, Stanford Research Institute, Palo Alto, Calif.

XUM

Over the years, the Camp Parks hot-cell facilities have developed and expanded largely by "cut and try" additions that satisfied certain specific requirements. Written operating instructions and shop drawings do not exist. The production and properties of this synthetic fallout and the facilities where it is made are briefly described here.

SYNTHETIC-FALLOUT-PRODUCTION PROCESS

The steps in producing synthetic fallout include (1) mineral processing to produce sized particles in ton quantities, (2) radioisotope processing in the hot cells, (3) radioisotope labeling of the mineral particles in concrete mixers and high-temperature furnaces, and (4) testing and control to ensure the desired radiochemical, chemical, and physical properties of the synthetic fallout.

Mineral Processing

Radioactive particles from 44 to 700 μ in diameter comprise a very large fraction of local fallout from a land-surface nuclear detonation. Four particle-size groups—44 to 88, 88 to 175, 175 to 350, and 350 to 700 μ—were produced to cover the common range.

Feldspar, quartz, and clay—the principal minerals in the earth's crust—were purchased in carload lots. Some of the minerals required crushing and pulverizing, but most were in the form of sand and required only sieving to produce the full range of particle sizes.

The particles were separated into sized groups on a commercial sieving machine, and fine particles were removed from the 44- to 88-μ material by a wet centrifugal method. Sieving efficiency was measured and controlled by frequent determinations of particle size. Each of the size groups was produced in ton quantities and stored in color-coded bags and barrels. The important physical properties of the four particle-size groups of one of the sands—Wedron—measured after careful sieving into a large number of intermediate sizes, were:

Size, μ	Number of particles per gram	Average surface area per particle, cm^2	Average particle diameter, μ
44–88	6.98×10^6	6.69×10^{-5}	47
88–175	6.20×10^5	3.21×10^{-4}	101
175–350	7.54×10^4	1.39×10^{-3}	210
350–750	7.09×10^2	7.85×10^{-3}	500

Radioisotope Labeling of Mineral Particles

Material is labeled with a radioisotope by spraying a weak acid solution of a selected radioisotope on a weighed charge of particles as they are tumbling in a rotating mixer. The particles are dried by direct heating or by introduction of heated air to the mixer. Subsequent treatment determines the solubility or availability of the radioisotope.

For a nonleachable synthetic fallout, the radioisotope is fixed on the mineral by an overcoat of sodium silicate, a solution of which is sprayed on the dry labeled particles while they are still in the mixer. The amount of sodium silicate is adjusted to produce a layer less than 1 μ thick. The labeled particles are redried, removed from the mixer, and placed in a furnace at 2000°F to fuse the sodium silicate layer and seal in the radionuclide. The physical properties of the mineral particles are not appreciably altered by this treatment. However, if a specified solubility of the radionuclide is desired, the labeled particles—without sodium silicate—are heated to a previously determined temperature that alters the particle surface and controls the combined chemisorption and diffusion of the radionuclide in the particle matrix.

The size of the batch being prepared dictates which of the available rotating mixers is selected for a particular operation. Ball mills and twin-shell blenders are used for gram and pound lots. Portable 1-ft^3 concrete mixers are used for lots of up to 100 lb, and specially modified 2-yd concrete mixers, which are charged by lift truck and hoppers, are used for 500-lb batches. After the radioisotope is sprayed on and dried and the sodium silicate is sprayed on and dried, the synthetic fallout is discharged on an endless belt that conveys it to a bucket elevator and metering hopper, where it is placed in stainless-steel pans. The pans are pushed by a hydraulic ram along skid rails into a gas-fired furnace. After 1 hr at 2000°F, the pans are pushed out the other end of the furnace for cooling. Further pushing automatically dumps the pans and discharges the material onto another endless belt for transfer to shielded hoppers. All these operations are performed from a remote and shielded location to minimize the radiation dose to personnel.

Testing and Control

All incoming shipments of radioisotopes and all outgoing batches of synthetic fallout are assayed with a 4π ionization chamber. The cylindrical steel chamber, which is argon-filled to 600 psig at 70°F, is 11 in. in diameter and 14 in. high, with a reentrant sample

XUM

thimble 1.75 in. in inside diameter by 12 in. deep. The entire chamber is shielded by 3 in. of lead. The useful ionization current ranges between 4×10^{-10} and 3×10^{-5} mA, and all readings are normalized to a standard response of 5.60×10^{-7} mA for 100 μg of radium.

The radiochemical purities of all incoming radioisotopes and all outgoing batches of synthetic fallout are verified using a gamma spectrometer with a 100-channel analyzer. The accumulated data are presented on a display oscilloscope and then read out on a paper-tape printer or an $X-Y$ plotter.

SYNTHETIC-FALLOUT FACILITIES

The synthetic-fallout facilities include equipment for mineral processing, testing and control, isotope preparation, and simulant production (Fig. 1). The area of greatest interest for this review is that for radioisotope processing. There are two hot cells, each with an inside floor area 8 by 8 ft. Shielding is provided by 2-ft-thick concrete walls, with a 2-ft-thick zinc bromide–filled viewing window. One cell is fitted with a pair of Model 8 Hevi-Duty Master-Slave manipulators and the other with a pair of Model 4 manipulators. Blowers maintain a slight negative pressure inside the cells, and about 500 cfm of leakage air is exhausted through absolute-filter banks. A 0.5-ton monorail hoist provides access to the cells and a shielded alleyway. One cell is equipped with a 0.25-ton jib crane that remains inside the enclosure. Each cell has through-wall holes for sample removal and for pressure, vacuum, and water lines and is supplied with a 100-A three-phase, four-wire electrical service. Work tables consist of stainless-steel trays atop cubic-yard concrete blocks that are carried on warehouse dollies. A 15-gal drum cast into the center of the concrete block receives waste for disposal. Separate work tables are set up for specific operations and are wheeled into the hot cell as needed. Solid waste is collected in polyethylene-lined drums and then transferred to approved shipping boxes. Liquid waste is poured into 5-gal polyethylene carboys and solidified with Micro-Cell E (a Johns-Mansville product). Ultimate disposal is contracted to a licensed company.

Small amounts of activity (up to a few millicuries) are processed in the chemistry laboratory, which is equipped with a hood and "hot" sink. Common items of laboratory apparatus and the most frequently used reagents are available so that a wide variety of qualitative and quantitative chemical tests can be conducted.

Radioisotopes that have been processed include kilocuries of ^{140}Ba–^{140}La, ^{147}Pm, and ^{204}Tl; multicuries of ^{85}Sr, ^{90}Sr, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{131}I, ^{137}Cs, ^{144}Ce, ^{177}Lu, and ^{198}Au; millicuries of ^{85}Rb and ^{134}Cs; and gross fission products. Many of these radioisotopes are produced by neutron irradiation in the nearby General Electric Test Reactor at Vallecitos.

The operation of the facility can best be illustrated by describing a single large production run and a small routine batch production run, made each month.

Large Production Run

One of the very large production runs required 10 tons of synthetic fallout labeled with 500 Ci of ^{140}Ba–^{140}La. Several acres were to be contaminated with 150- to 300-μ particles at a surface density of 90 g/ft^2, and an initial gamma radiation field of 100 mR/hr at 3 ft above the surface was required for instrument response.

Kilocurie quantities of ^{140}Ba–^{140}La were to be used. This radionuclide mixture was selected because the 12.8-day half-life of the parent was adequate for the test period and yet short enough to create no long-term contamination problem, its broad spectrum of gamma energy was adequate for good instrumentation, and its chemical properties ensured that it could be fixed to the sand particles. In addition, ^{140}Ba–^{140}La is found in fallout and contributes about 80% of the gamma radiation emitted from 20-day-old fission products, and approximately 1 mCi/ft^2 of ^{140}Ba–^{140}La uniformly deposited on an essentially infinite plane yields 100 mR/hr at 3 ft above the surface. It was calculated that ten 500-lb batches of synthetic fallout would be required and that each batch would be labeled with 50 Ci of ^{140}Ba. Ten batches of synthetic fallout were produced on schedule and without incident. Radioassay showed a very uniform specific activity from batch to batch.

To minimize the radiation dose to personnel, the sand was labeled with only the weak-gamma-emitting parent ^{140}Ba. About 90% of the potential radiation dose from an equilibrium parent–daughter mixture was avoided by precipitating barium nitrate and discarding the nitric acid solution containing the daughter ^{140}La. Storage of the synthetic fallout for 10 days allowed the ^{140}La to grow and reestablish the equilibrium.

Monthly Batch Runs

Every month a batch of sand was labeled with ^{90}Y. From a 30-Ci ^{90}Sr generator, ^{90}Y is recovered by

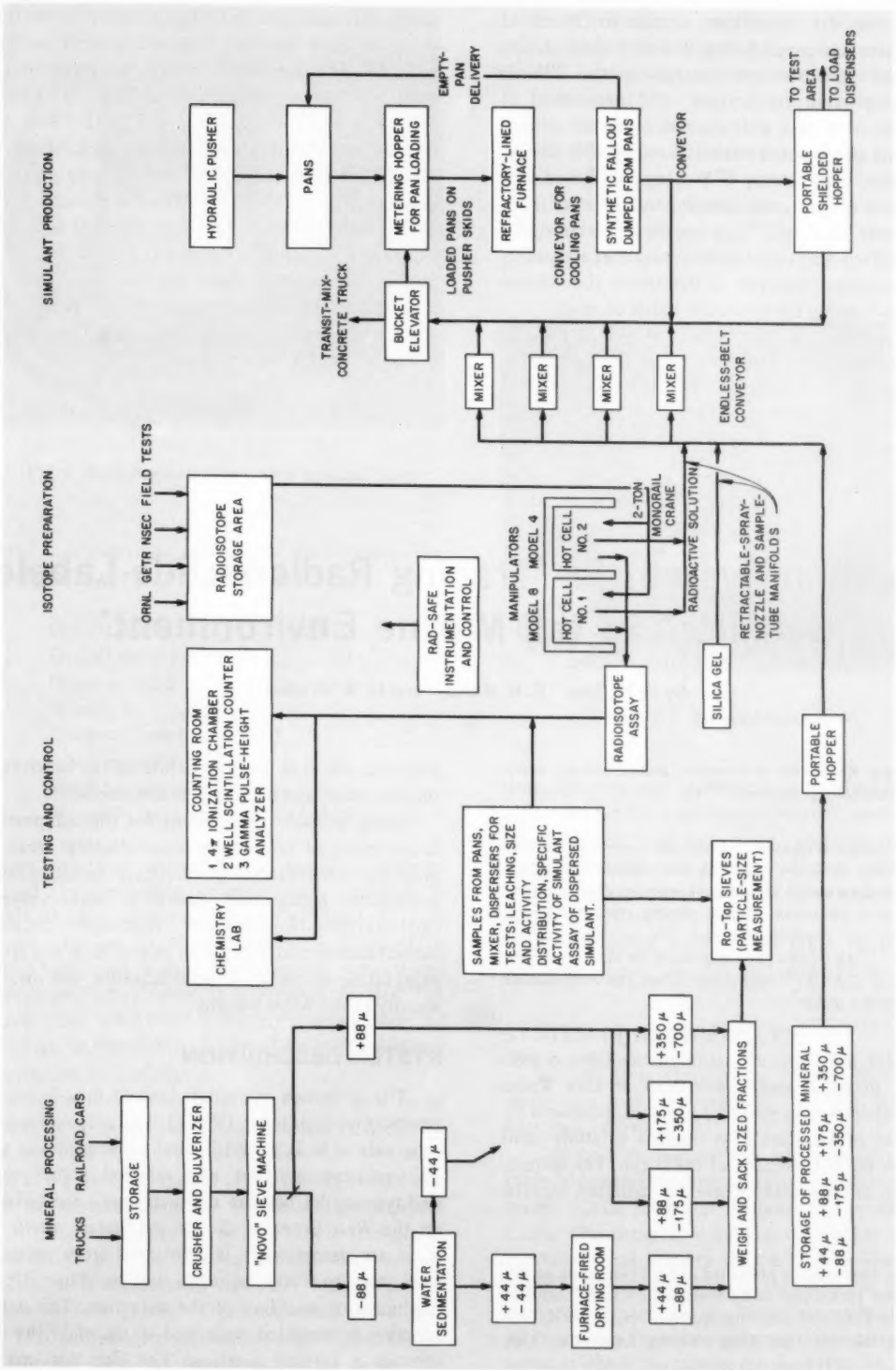

Fig. 1 Flow sheet for preparation of synthetic fallout.

dissolving the dry strontium nitrate in 25 ml of distilled water and precipitating it with 125 ml of 90% HNO_3. The acid solution containing the ^{90}Y is filtered, evaporated to dryness, and redissolved in 25 ml of water. Then 2 g of inactive strontium nitrate is added and precipitated with 125 ml of 90% HNO_3, and the acid solution of ^{90}Y is again filtered off, evaporated to dryness, and dissolved in 100 ml of 0.1*M* HNO_3. About 20 Ci of ^{90}Y is usually available at this point. A 100-μl aliquot of this solution is assayed in the 4π ionization chamber to determine the volume required for labeling the particular batch of sand.

Sufficient Wedron sand to meet the batch requirements is prepared by wet-sieving and Ro-Tapping to ensure that all particles are within the specified size range. The sand (up to 600 g) is added to the rotating drum of a ball mill that is operating in the second hot cell, and the calculated volume of carrier-free ^{90}Y solution is sprayed on the tumbling particles. The radioisotope-labeled sand is dried by the heat from a hot plate placed directly under the metal drum. Then 10 ml of sodium silicate is sprayed into the rotating drum to coat the particles. After the particles are again dried, the synthetic fallout is transferred to a crucible and placed in a muffle furnace at 1950°F for 1 hr. The product is removed from the furnace, cooled, and returned to the hot cell for assay. When the specific activity is within acceptable limits, the synthetic fallout is packaged and shipped.

(MG)

Detection System for Tracing Radionuclide-Labeled Sediment in the Marine Environment*

By F. N. Case, E. H. Acree, and H. R. Brasheart†

Supplementary Keywords: *instrument; sedimentology; material tracing; oceanology; mobile;* ^{198}Au.

Abstract: *A system is described which can trace radionuclide-labeled sediment on a dry beach, in the breaker zone of the ocean, and in deep water. The sodium-activated CsI detector is transported in a wire-mesh-covered rotating container that is towed behind an amphibious vessel. The sensitivity of the detector for* ^{198}Au*-labeled sand deposited on the bottom of a water body is* 2.3×10^3 *counts/sec when the radionuclide concentration is 1* $\mu Ci/ft^2$.

A system for use in tracing radionuclide-labeled sediment was designed and constructed at Oak Ridge National Laboratory as part of a program sponsored by the Division of Isotopes Development to study sand transport in the littoral zone of the ocean. The system, designed to automatically correlate radiation counts, position, and time and to operate on the beach face, in the surf zone, and off shore, is described here.

Other possible applications for the equipment are in surveying of underwater waste-disposal areas, mapping the distribution of dredging spoils, obtaining underwater background surveys in areas subject to radioactive-process-effluent discharge, conducting natural-background surveys in inland lakes and streams, prospecting for heavy mineral deposits, and measuring activity in the water column.

SYSTEM DESCRIPTION

The detection system is housed in a heavy wire-mesh-covered cylinder (Fig. 1) that rotates around its long axis as it is towed behind an amphibious vessel. The rotating cylinder was selected rather than a sled-type trailer because the instrument was to be used on the West Coast of the United States, where rocky areas are common. It is fabricated from rectangular steel bars to form an open lattice. This affords a minimum of shielding of the detectors. The detector chamber is weighted with lead to maintain the detectors in a vertical position. The detector units are

*Extracts from a paper presented at the Symposium on the Use of Nuclear Techniques in the Measurement and Control of Environmental Pollution, Salzburg, Austria, Oct. 29, 1970.

†Isotopes Division, Oak Ridge National Laboratory, Oak Ridge, Tenn.

XUM

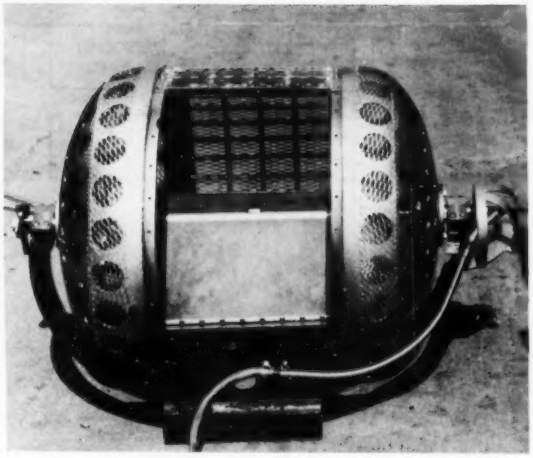

Fig. 1 Radiation-detection-system housing.

mounted in a watertight chamber of the cylinder. The physical characteristics of the cylinder are

Overall width, in.	50
Overall diameter, in.	30
Housing width, in.	43
Height, in.	30
Distance from center shaft to tow point, in.	32
Weight in air, lb	505
Weight in fresh water, lb	410

Four radiation detectors—2- by 2-in, sodium-activated cesium iodide crystals, each enclosed in 0.030-in.-thick aluminum (Fig. 2)—are used. Because they are exposed to water pressure, a 0.5-in.-thick Plexiglas light pipe is placed between the crystals and the photomultiplier tubes as a pressure barrier. The system can detect 2.3×10^3 counts/sec of ^{198,199}Au-labeled sediment spread on the littoral at a concentration of 1 μCi/ft^2.

The detector system is towed on the area to be scanned by a 60-ft amphibious vehicle (LARC-15), which can operate in 6- to 8-ft breakers. The cable that connects the detector system to the towing vehicle also carries the detector signals to the recording equipment (Fig. 3). This coaxial cable, with a tensile strength of 30,000 lb, is part of a wire bundle that has an additional nine conductors that carry high voltage to the photomultiplier tubes from individual power supplies. This permits gain adjustment of the tubes so that the responses from the four detectors are equal. The preamplifier output is amplified with a linear amplifier, which feeds two differential discriminators, making it possible to select two segments of the gamma spectrum to be recorded. The discriminator output signals drive two buffered scalers, which are interrogated by a master control unit that records the data on an 8-level punched paper tape. The print-out control unit also interrogates scalers, which generate a line number and a time record, and a range finder to establish boat position. This provides an automatic correlation of

Fig. 2 Detector components.

time, location, and radiation counts. The boat position is determined by a microwave system, which indicates the distances from two fixed positions on shore. A complete set of data can be collected every 1, 2, or 5 sec or multiples of 10 of any of these times by switch selection on the console.

The punched tape is the input to a computer, which interprets the data and prepares contour maps.

SYSTEM PERFORMANCE

In field tests, the rotating housing moved satisfactorily over rocks that were up to 2 ft in diameter and over rock outcrops.

During a normal survey, approximately 100 ft of cable attaches the detector-system vehicle to the tow vessel, which moves at a speed of approximately 3 knots. The detector vehicle is very stable under these conditions and remains on the bottom. The length of cable required to maintain the detector vehicle in a surveying position on the littoral depends on the depth of the ocean floor beneath the water surface and the speed at which the system is towed (Fig. 4).

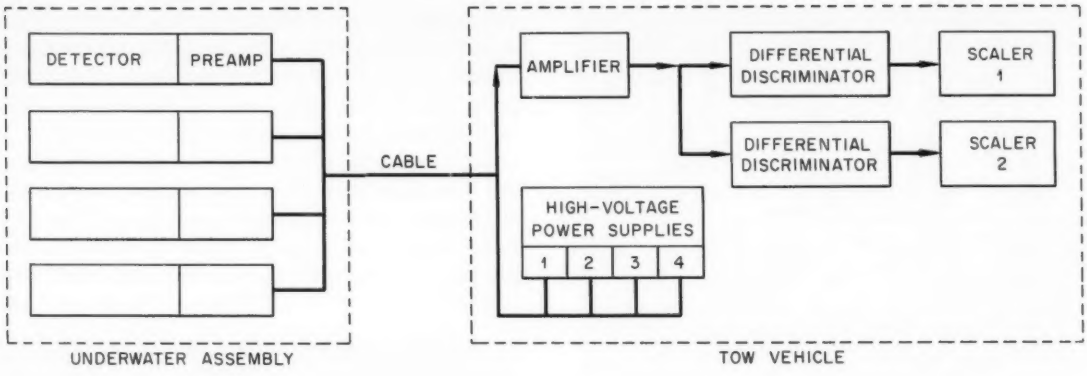

Fig. 3 Schematic of underwater and surface equipment used in tracing radioisotope-labeled sand in the marine environment.

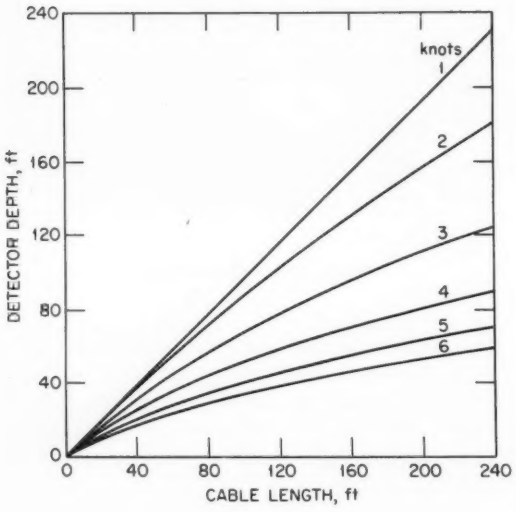

Fig. 4 Minumum cable length for satisfactory operation of detector vehicle at various depths and speeds.

In a study of a silting harbor at Oceanside, Calif., in February and March 1970, excellent radiation profiles, representing sediment transport into the harbor and around the protective structures, were obtained in a location where fluorescent-tracer experiments had failed. The harbor is protected by breakwaters to the northwest and south, and background radiation counts were variable outside the breakwater. This variation, which is characteristic of the West Coast of the United States, is due to deposits of thorium-containing black sand. A line injection of labeled sand was made outside this breakwater north of the harbor outlet and inside the harbor perpendicular to the beach, and two point injections were made 5 days later, one at the northern edge of the harbor inlet and the other outside the harbor inlet. Radiation contours showed that sand in the harbor was transported into the harbor inlet and from the beach into the ship channel, i.e., to areas where shoaling occurs and makes dredging necessary to maintain navigation channels.

(MG)

Section

II

Isotopes and Radiation Technology

Process Radiation Development

Editors' Comment: One of the most controversial, yet potentially vast applications of process radiation is the treatment of sewage and other waste waters. Some reseachers have proposed treatments that are claimed to be economically competitive with existing methods; others say "not so." Inconsistencies exist in reported bacterial kill factors, efficiencies of sedimentation and oxidation, preferred method of treatment, etc. Rather than to try to evaluate these proposals (which might be rather ill-advised at this time), we are including them "on their own merits" in order to present, as completely as possible, the present state of the art, uncertain as it may be.

Any questions or comments should be directed to the individual authors concerned.

Potential Role of Radiation in Waste-Water Treatment*

By D. S. Ballantinet

Supplementary Keywords: *pollution; water and waste treatment; industry, utility; radiolysis; economics; source, gamma; X-ray machine; source, beta; accelerator; modification, material; microbiology; sterilization.*

Abstract: *The potential role of radiation in the treatment of waste water is described. Interactions of radiation with both water and organic compounds are briefly presented, and dose requirements are discussed. The use of radiation to concentrate solids and destroy bacteria is also considered. Irradiation appears to have some attractiveness in increasing the sedimentation rate of solids; however, low reaction efficiencies of radiation in the destruction of waste materials (G values < 10) indicate a cost greater than that for conventional technology in the treatment of sewage.*

The concept of using nuclear radiation for sewage treatment goes back to the 1950s, but, during the past several years, both the semitechnical and nontechnical press have frequently indicated interest in this application. This article reviews the characteristics of nuclear radiations and their interactions with aqueous and organic systems with the hope of placing experimental work—completed and in progress—in a rational framework.

A review of problems of waste-water management indicates that major concerns are with (1) the reduction of biological oxygen demand (BOD) and chemical oxygen demand (COD), (2) the conversion of refractory compounds to a form more readily oxidizable by biological or chemical agents, (3) the destruction of bacteria, and (4) the improvement in handling and disposal of solids. We may now ask, "Can and how may radiation be useful in attacking these problems?"

The term "nuclear radiation" includes neutrons, alpha particles, heavy charged particles from ion accelerators, accelerated electrons, beta particles, gamma rays, and X rays, but some of these forms of radiation can be eliminated from serious consideration. Neutrons are undesirable because they can induce

*This paper was presented at the 62nd Annual Meeting of the American Institute of Chemical Engineers, November 16–20, 1969, Washington, D. C., and is published here by permission.

†Division of Isotopes Development, U. S. Atomic Energy Commission, Washington, D. C.

XUM

radioactivity readily in municipal or industrial wastes. Alpha and other heavy charged particles have extremely limited penetration capabilities and would be totally ineffective in any conceivable waste-water-treatment method. The remaining forms of radiation, which can be seriously considered, may conveniently be grouped into two classes: (1) beta particles and accelerated electrons and (2) gamma rays and X rays. The distinction is the greater penetrating capability of the gamma rays and X rays.

RADIATION-INITIATED REACTIONS

Let us now consider how these particles and radiations interact with matter, recognizing that the interactions by the two classes are similar. The sequence of reactions

$$AB + \gamma\ (\text{or } e^-) \rightarrow AB^* \quad \text{and} \quad AB^+ + e^- \qquad (1)$$

$$AB^+ + e^- \rightarrow AB^* \qquad (2)$$

$$AB^* \rightarrow A\cdot + B\cdot \qquad (3)$$

$$AB+ \rightarrow A^+ + B\cdot \qquad (4)$$

indicates that the initial process (reaction 1) is one of electronic excitation and ionization of the target molecule, AB. Subsequently, electronic excitation can also be produced as a consequence of the ion recombination (reaction 2). The electronically excited AB* molecule can fragment into two free-radical species, A· and B· (reaction 3), and the AB+ ion can fragment to an A^+ ion and a free radical, B·. This description is a great oversimplification, but it indicates that these processes can cause fragmentation of the target molecule and produce highly reactive ions and free radicals. In a waste-water-treatment context, these processes can be viewed either as fragmentation or as an increase in the chemical potential of a pollutant molecule.

The action may be either direct or indirect. Direct action may be considered as an interaction of the nuclear radiation with a target molecule, while indirect action occurs when the target molecule is affected by some intermediate chemical species activated or formed as a result of radiation interaction. Obviously, in pure compounds, all effects result from direct action, but, in solutions, indirect action will usually be the principal mechanism. For example, a waste water may contain about 1% organic matter—perhaps sugar molecules. The probability of a direct interaction of the radiation with a sugar molecule is small since most of the radiation will be absorbed by the solvent, water. Any significant radiation effect in waste water must relate to the interaction of the radiation with water molecules and the species generated by this interaction.

Fortunately, aqueous radiation chemistry has commanded the attention of the most eminent radiation chemists throughout the world for a quarter of a century. Today, it gives not only quantitative data on the yield of radiolysis products but also rate constants for the reaction of these radiolysis products with many inorganic and organic solute molecules.[1]

Interactions with Water

Very briefly, the interaction of radiation with water leads to the reactions

$$H_2O \rightarrow H_2O^* \qquad (5)$$

$$H_2O^* \rightarrow H\cdot + \cdot OH$$

and

$$H_2O + \rightarrow H_2O^+ + e^- \qquad (6)$$

$$e^- + aq \rightarrow e^-_{aq}$$

$$H_2O^+ + H_2O \rightarrow H_3O^+ + \cdot OH$$

in which are formed the reducing species H·, the hydrated electron e^-_{aq}, the oxidizing species ·OH, and the oxidizing molecular product H_2O_2. As indicated earlier, the yields of these various products have been accurately determined[2] as follows:

$$H_2O \rightarrow e^-_{aq}(2.6);\ OH(0.6);\ H(0.5);\ H_2O{+}(3.6);\ H_2O_2(0.7);\ OH^-(1.0);\ H_2(0.45) \qquad (7)$$

where the numbers in parentheses are the G values (the number of molecules or species formed or decomposed, or the number of chemical events, resulting from the absorption by the system under study of 100 eV of energy). Thus, in pure water, for each 100 eV of energy absorbed, 2.6 OH radicals, 0.5 H radicals, and 0.7 H_2O_2 molecules, etc., are formed. Actually, most waste water contains dissolved oxygen, with which both H· and e^-_{aq} can react to form an additional oxidizing species, O_2^- :

$$H\cdot + O_2 \rightarrow HO_2\cdot \qquad (8)$$

$$HO_2\cdot \rightarrow H^+ + O_2^-$$

and

$$e^-_{aq} + O_2 \rightarrow O_2^- \qquad (9)$$

Returning now to the possible utility of radiation in waste-water treatment, we can attempt to develop

XUM

some semiquantitative ideas for possible decrease in COD or destruction of organic solutes and for altering refractory components. For any given situation, the concentration of the pollutant of concern, the G value for some chemical change of the pollutant, and the amount of radiation energy which is delivered for the treatment must be known.

Interactions with Organic Compounds

We can optimistically assume that each of the primary species H·, ·OH, and e^-_{aq} has the capability of attacking and altering an organic molecule. The attack of a hydrocarbon molecule by H· to yield a hydrocarbon radical and a molecule of hydrogen gas may be represented by

$$RH + H\cdot \rightarrow R\cdot + H_2 \qquad (10)$$

where a hydrocarbon molecule is attacked by a H· atom to yield a hydrocarbon radical and molecule of hydrogen gas. The fate of this particular radical depends on its reactivity, its concentration, and the relative reactivity and concentration of other free-radical species or components of the mixture capable of reacting with free radicals; e.g., one can visualize the reactions

$$R\cdot + H\cdot \rightarrow RH \qquad (11)$$

$$R\cdot + R\cdot \rightarrow RR \qquad (12)$$

$$R\cdot + R^1\cdot \rightarrow RR^1 \qquad (13)$$

$$R\cdot + \cdot OH \rightarrow ROH \qquad (14)$$

$$R\cdot + O_2 \rightarrow RO_2\cdot \qquad (15)$$

Reaction 11 represents a simple recombination with no net change; reactions 12 and 13 represent a combination of two hydrocarbon radicals, similar in reaction 12, dissimilar in reaction 13. Reaction 14 can be viewed as a simple one-step oxidation, and reaction 15 the special case of oxidation by dissolved molecular oxygen. That an organic molecule is not really destroyed in any of these reactions is fairly obvious. Reaction 11 re-forms the parent molecule, and reactions 12 to 14 yield a new organic molecule of somewhat different composition. The new molecules may be more readily attacked by bacteria so that a net reduction in BOD may eventually be achieved. This possibility lends some credence to the postulation that radiation can produce a more easily biodegradable product. Conversely, the product molecule may also be more resistant to biodegradation than the parent.

Dose Requirements

Let us assume that the refractory target pollutant has a molecular weight of 100 and that each of the radiolysis products—H·, G(0.5); ·OH, G(2.6); and e^-_{aq}, G(2.6)—converts this compound to a more readily biodegradable compound. The total yield of these species is 5.7. For a dose of 100 thousand rads,* 6.2×10^{18} eV/g or about 6.2×10^{21} eV/liter will be absorbed. Calculation shows that this dose under the most ideal conditions will convert 60 mg/liter of the refractory target material with a molecular weight of 100 to the more readily biodegradable form:

$$\frac{6.2 \times 10^{21}\ \text{eV}}{\text{liter}} \times \frac{5.7\ \text{molecules}}{100\ \text{eV}} \times \frac{100{,}000\ \text{mg/mole}}{6 \times 10^{23}\ \text{molecules/mole}} = \sim 60\ \text{mg/liter}$$

Some reasonable estimates of the efficiency of radiation in decreasing the COD can be made. Among the radiolysis products, only ·OH (G = 2.6) is directly an oxidant (·OH → OH^-), but, in the presence of dissolved oxygen, we may also anticipate

$$e^-_{aq} + O_2 \rightarrow O_2^- \qquad (16)$$

$$H\cdot + O_2 \rightarrow HO_2\cdot \qquad (17)$$

$$HO_2\cdot \rightarrow H^+ + O_2^-$$

where the H· and e^-_{aq} species are also converted to the oxidant O_2^-. Each O_2^- is capable of contributing three additional oxidation events; thus, in the ideal situation, where each primary species is involved in oxidation, we can expect a G of about 12 [i.e., $(G_{e^-} + G_{H\cdot}) \times 3 + G_{\cdot OH}$].

Also, the dissolved molecular oxygen may be involved in oxidative processes:

$$R\cdot + O_2 \rightarrow RO_2\cdot \qquad \text{initiation}$$

$$\left.\begin{aligned} RO_2\cdot + RH &\rightarrow RO_2H + R\cdot \\ R\cdot + O_2 &\rightarrow RO_2\cdot \\ RO_2\cdot + RH &\rightarrow RO_2H + R\cdot \end{aligned}\right\} \quad \text{propagation}$$

$$R\cdot + \cdot OH \rightarrow ROH \qquad \text{termination}$$

Such a sequence appears to be involved in long oxidative chains, but actual G values for a number of

*One rad is equivalent to the absorption of 100 ergs of radiation energy per gram of material, or 6.2×10^{13} eV/g.

organic compounds[3] have been found to be <10. One obvious limitation is the availability of dissolved oxygen, which at 20°C is about 8 mg/liter. If we assume a G value of 10 (here G refers to the number of 1-electron reactions per 100 eV), which seems reasonable from a consideration of aqueous radiolysis and actual experimental data,[3] a dose of 100 thousand rads will produce 6×10^{20} oxidations per liter or approximately 1×10^{-3} equivalent of oxidant per liter. A molecule of oxygen is capable of giving four oxidation events, which is equivalent to 0.25 value of O_2 per liter or about 8 mg/liter. These values clearly indicate that any process in which a ΔCOD of even 10 mg/liter is envisioned will involve reaeration.

Another way of evaluating radiation requirements is to assume some target ΔCOD and G value for the oxidation. A very simple equation gives

$$\text{rads} = \frac{\Delta\text{COD}}{\text{G}} \times \frac{6 \times 10^{23}}{8 \times 10^{3}} \times \frac{10^{2}}{6.2 \times 10^{16}}$$

where ΔCOD = milligrams per liter
6×10^{23} = Avogadro's number
8×10^{3} = equivalent weight of oxygen (as an oxidant), mg
10^{2} = eV
6.2×10^{16} = number of electron volts absorbed per liter per rad
G = G value

This equation can be simplified to

$$\text{rads} = \frac{\Delta\text{COD}}{\text{G}} \times 1.21 \times 10^{5}$$

Thus, if one desires a ΔCOD of 100 and the G value is 10, a dose of 1.2×10^{6} rads is required.

Destruction of Pollutants

An examination of the literature is helpful in assessing the practicability of radiation treatment for specific classes of pollutants. Table 1 gives G values for disappearance in aqueous solutions of representatives of various classes of organic compounds—some experimentally determined and others estimated from the G values of formation of different radiolysis products.

We can also estimate the rate at which various organic pollutants may be expected to react with the initial aqueous radiolysis products. Rate constants for reactions of many organic compounds have been measured by pulse-radiolysis techniques (Table 2). There are some obvious dangers in trying to extrapolate these kinetic data from pure aqueous solutions to the highly complex municipal waste-water streams in which a wide spectrum of organic components, some unidentified, would be engaged in simultaneous competitive reactions. Even in complex mixtures, some measure of the gross reaction rate of the pollutants can

Table 1 G Values for Disappearance of Organic Compounds in Air-Saturated Aqueous Systems

Compound	G (−)*	Remarks
Tyrosine	1.8–3.6	Over *p*H range 1–8
Phenyl alanine	2–7	*p*H 1–8
Serine	1.5–5.5	G increases with increasing concentration
Cysteine	7–9.3	*p*H 0.5, 5.6
Diphosphopyridine nucleotide	6.9	
Glucose	1.5–3.5	$5.5 \times 10^{-4} M$ to $5.5 \times 10^{-2} M$
Methyl glucoside	3.2	
Phenyl glucoside	3.1	
Uracil	12.5	
Dimethyl uracil	3.15	*p*H 7
Phenol	0.5	
α-Amino benzoic acid	0.99	
Benzoic acid	2.4	*p*H 8
p-Nitrophenol	4.3	
Formic acid	4.1	
Oxalic acid	2.9	
Succinic acid	1.1	
Methylene blue	3.4	
Citral	4.8	
Citronellal	6.3–6.8	

*Number of molecules destroyed for each 100 eV of radiation energy absorbed.

Table 2 Bimolecular Rate Constants of Radicals with Organic Compounds in Aqueous Solution

Compound	$k \times 10^{-10}$ moles/sec		
	e^-_{aq}	H·	·OH
α-Alanine	0.0005		0.005
Aniline	0.002	0.18	0.53
Benzoate ion	0.31	0.087	0.33
Chloroform	3.0	0.000085	0.00085
Cystine	1.3	70.15	0.32
Ethanol	< 0.00001	0.0016	0.12
Glucose	< 0.0001	0.004	0.1
Phenol	< 0.0004	0.42	0.42
Urea	0.00003		< 0.00007

XUM

be determined by competitive pulse-radiolysis techniques. For example, phenol or some other compound that reacts rapidly and at a known rate can be added in known quantities for determination of its ability to compete against gross contaminants. By suitable selection of known additives, gross rates for reaction with H•, •OH, and e^-_{aq} can be determined.

In summary, there are numerous data on basic aqueous radiation chemistry that are useful in estimating the potential efficiency of radiation in producing changes in organic materials. Data obtained in very pure systems suggest that efficiencies are low, with G values considerably less than 10. While oxidation could, in principle, be an efficient reaction, the data indicate that G values are generally only about 10 or less.

SEDIMENTATION OF SOLIDS

The sediment streams from primary and secondary treatments generally contain several percent solids, which must be concentrated by any of a variety of techniques—e.g., filtration, flocculation, and distillation. Irradiation might be capable of increasing the sedimentation rate or altering the colloidal character of solids so that they could be more readily dewatered and would remain sufficiently mobile to be pumped at higher concentrations. Such improvements would materially decrease the size of waste-solids handling systems and increase the capacity of existing plants.

The stream in which solids have been concentrated—the sludge—generally makes up only 1% of the total volume flow through waste-water-treatment plants; yet 50% of the total cost of waste-water treatment is related to sludge handling. Both these factors increase the attractiveness of irradiation as a sludge-treatment technique.

Unfortunately, there has been little critical work done on irradiation of highly colloidal waste waters to alter the sedimentation rate and increase the solids concentration. In general, colloids are stabilized by electrostatic charges and by insulating shells. The electrostatic charges cause mutual repulsion between particles and prevent their aggregation to a size sufficient for gravity forces to cause sedimentation.

In principle, the use of radiation to cause decrease or destruction of the electrostatic forces and alteration of the insulating shell is feasible. Irradiation of aqueous systems produces ions and free electrons, and these could react with oppositely charged species and neutralize the charge on colloidal particles in the way that addition of electrolyte neutralizes charges and causes subsequent sedimentation. However, classical studies indicate that the effect of added electrolyte is dependent on concentration and may either increase or decrease the stability of the colloidal systems.

Since radiation can produce changes in organic materials, reduction of the hydrophilic character of solids by radiation may conceivably be possible. Such a change should decrease the shell effect and thus increase the sedimentation rate. However, the opposite is equally possible, and indications are that the radiation effect is usually unpredictable and of limited magnitude.

DESTRUCTION OF BACTERIA

The use of radiation for destruction of bacteria has been extensively studied in other areas of technology—e.g., pasteurization and sterilization of food products, sterilization of medical supplies, pasteurization of enzyme-containing soap products, and sterilization of foot powders. The sterilizing dose is generally accepted as 2.5 Mrads, which will handle the most resistant species, the spore-forming organisms.

In assessing the potential of radiation in waste-water treatment, it is essential to first establish criterions for acceptability, including not only the percentage kill but also the maximum permissible residual populations of specific bacterial strains that may be released into the environment. A recent report[4] claims 99.9% kill for *Escherichia coli* but only 95% for enterococci in municipal waste water with a dose of 100 thousand rads and indicates that the enterococci value is the more significant. Obviously, the significance of 95% kill for an initial population of 1 million is quite different from that where the initial population is 10-fold higher or lower.

Although alternative methods using chlorine and ozone are effective for killing bacteria, the penetration capability of radiation makes it more effective for bacteria retained within particulates.

The combination of radiation and chlorine may lead to a synergistic effect in bacterial kill in waste water, such as the kill of bacteria in drinking water[5] (chlorine demand free). Likewise, a synergistic effect has been reported[6] for the combination of chloroform and X radiation to inactivate bacteriophage.

Various means, both physical and chemical, for increasing the effectiveness of radiation in killing bacteria have been reported. Heat combined with radiation showed[7] a two-orders-of-magnitude synergistic effect, and prior ultrasonic treatment has been shown[8] to sensitize microorganisms to radiation. Costs

associated with each member of the synergistic combination must be considered, and the resultant magnitude of the effect must be large enough to justify those costs.

In all these potential radiation applications, cost is the most important consideration—particularly if the total throughput of a plant must be treated. In spite of optimistic predictions to the contrary, a realistic figure for radiation-treatment costs is 1 cent per megarad-pound. Thus, if waste water requires a dose of 1 Mrad to achieve the desired objective, the cost will be \$83 per 10^3 gal. Even if the optimistic predictions of achieving a decrease in radiation-dose requirements to 100 thousand rads and a reduction of radiation costs by a factor of 10 should be realized, the treatment costs will still be \$0.83 per 10^3 gal. Against this we have the statements[9] of the Federal Water Quality Administration, formerly the Federal Water Pollution Control Administration, that existing proven technology can completely renovate municipal waste water to drinking-quality water for about \$0.60 per 10^3 gal.

Hopefully, research on the radiation treatment of waste water will permit more accurate evaluation of the economics and the delineation of those areas where additional research is warranted. (REG)

REFERENCES

1. M. Anbar and P. Neta, A Compilation of Specific Bimolecular Rate Constants for the Reactions of Hydrated Electrons, Hydrogen Atoms and Hydroxyl Radicals with Inorganic and Organic Compounds in Aqueous Solution, *Int. J. Appl. Radiat. Isotop.*, **18:** 493-523 (1967).
2. A. O. Allen, *The Radiation Chemistry of Water and Aqueous Solutions*, D. Van Nostrand Company, Inc., Princeton, N. J., 1961.
3. D. M. J. Compton, S. J. Black, F. L. Lieurance, and W. L. Whittemore, Application of Ionizing Radiation to the Treatment of Waste Waters and Sewage Sludge, in *Large Radiation Sources for Industrial Processes*, Symposium Proceedings, Munich, 1969, pp. 399-416, International Atomic Energy Agency, Vienna, 1969 (STI/PUB/236).
4. C. J. Touhill, E. C. Martin, M. P. Fujihara, D. E. Olesen, J. E. Stein, and G. McDonnell, The Effects of Radiation on Chicago Metropolitan Sanitary District Municipal and Industrial Wastewater, *J. Water Pollut. Contr. Fed., Res. Suppl.*, **41**(2): R44-R60 (1969).
5. G. I. Sidorenko, M. A. Pinigin, and V. T. Savenkova, Use of the Combined Action of Gamma Radiation and Chlorine on Water Sterilization, *Gig. Sanit.*, **28**(9): 97-98 (1963); English Translation, Report JPRS-21, 975, pp. 10-12.
6. I. M. Cotton and L. S. Lockingen, Inactivation of Bacteriophage by Chloroform and X Irradiation, *Proc. Nat. Acad. Sci.*, **50:** 363-367 (1963).
7. G. Stehlik and K. Kaindl, Microbiological Studies on the Influence of Combined Processes of Heat and Irradiation on the Survival of Saccharomyces Cerevisiae var. Ellipsoideus, in *Food Irradiation*, Symposium Proceedings, Karlsruhe, 1966, pp. 299-305, International Atomic Energy Agency, Vienna, 1966 (STI/PUB/127).
8. S. D. Dharkar, Sensitization of Microorganisms to Radiation by Previous Ultrasonic Treatment, *J. Food Sci.*, **29:** 641-643 (1964).
9. L. W. Weinberger, D. G. Stephan, and F. M. Middleton, Solving Our Water Problems—Water Renovation and Reuse, *Ann. N. Y. Acad. Sci.*, **136** (Art. 5): 131-154 (1966).

High-Energy Radiation Synthesis of Ozone for Water Treatment*

By Meyer Steinberg and Morris Bellert

Supplementary Keywords: *reactor; source, beta; source, gamma; industry, chemical; industry, utility; microbiology; water and waste treatment; sterilization; synthesis; pollution; pilot-plant scale; cobalt-60; strontium-90; cesium-137.*

Abstract: *A radiation process for producing ozone from atmospheric oxygen has been designed on the basis of the results of radiation-chemistry studies and operation of the Brookhaven in-pile research loop as a pilot plant. Ozone for treating sewage or drinking water can be produced in a high-pressure facility with yields at least two to three times those obtained with the low-pressure electric-discharge ozonizer process. The fission-fragment chemonuclear process has a large potential for producing hundreds of tons of ozone per day for treating sewage-plant secondary effluent volumes of 10^8 to more than 10^9 gal/day. At these rates the cost of ozone, using municipal fixed-charge rates, can be as low as 2*

*Summary of USAEC Report BNL-13812, July 1969. The work, performed under the auspices of the U. S. Atomic Energy Commission, Division of Isotopes Development, was reported at the 62nd Annual Meeting of the American Institute of Chemical Engineers, Washington, D. C., Nov. 16–20, 1969.

†Brookhaven National Laboratory, Upton, N. Y. 11973.

XUM

cents per pound, which is about half that of producing ozone by the electric-discharge process. The use of radioisotopic radiation, such as that from the long-lived waste fission-product ^{137}Cs, may be considered for treating small quantities of secondary sewage effluent—<10^7 gal/day—or for drinking-water disinfection.

Ozone, which has one of the highest oxidation potentials in aqueous solution of any of the commonly used oxidants, has been effectively used for water treatment. For treatment of the residual refractory organics in sewage-plant secondary effluent, ozone treatment competes with the process of adsorption on activated carbon. For drinking-water purposes, ozone competes with chlorine.

When used to treat effluents from waste-water-treatment plants,[1-3] ozone lowers the chemical oxygen demand (COD) and total organic carbon (TOC), destroys pathogenic organisms, and removes odors and colors. Secondary sewage from a treatment plant typically contains between 70 and 150 ppM COD, which can be decreased to about 35 ppM by coagulation treatment. Since good-quality surface water has a COD of less than 15 ppM, this secondary effluent should be treated to decrease the residual COD by at least 20 ppM. The ozone concentration needed for such a decrease in COD, assuming an 80% utilization efficiency and using one effective O atom per O_3 molecule, is thus 3 x 20/0.8 or 75 ppM O_3 by weight of the sewage effluent. Hence approximately 32 tons of ozone per day would be required to treat a sewage-plant effluent of 10^8 gal/day.

When ozone is used in treating drinking water, much lower concentrations are needed—e.g., 2 ppM—in which amounts it could compete with chlorine. The product has no residual odor or unpleasant taste.

The major drawback to the general use of ozone for treating sewage-plant effluent or drinking water has been the cost of producing it. The only known process for producing commercial quantities of ozone is the electric-discharge ozonizer process, in which an a-c corona discharge is produced across a glass dielectric between two electrodes at a potential of 50 kV. Oxygen separated from the air or air itself is passed through the discharge, and the molecular oxygen is decomposed to O radicals, which unite to form ozone. The unit operates at atmospheric or a few pounds above atmospheric pressure. Ozone concentrations of 2 to 4% are obtained with oxygen feed. A single unit consists of a number of glass-lined discharge tubes in an assembly similar to a heat exchanger. Since the largest single unit made produces about 0.5 ton/day, a number of units would be required for treating large quantities of sewage.

BASIC ENERGY REQUIREMENTS

The lowest cost raw material for producing ozone is oxygen from the atmosphere. The synthesis of ozone from oxygen is an energy-dependent endothermic process, requiring 34 kcal of energy per mole of ozone,

$$\tfrac{3}{2}O_2(\text{gas}) \rightarrow O_3(\text{gas}) \qquad \Delta H_{298.2} = 34 \text{ kcal/mole}$$

and the efficiency of a process for production of ozone is shown by its energy requirement compared to this minimum value (Table 1). The electric energy required for the electric-discharge ozonizer process ranges[4,5] from 3.75 to 5.0 kW(e)-hr/lb. This energy is equiva-

Table 1 Yields and Energy Requirements for Ozone Synthesis

$\tfrac{3}{2}O_2(\text{gas}) \rightarrow O_3(\text{gas}) \qquad \Delta H_{298.2} = 34.0$

Maximum G Value = 68.1 molecules/100 eV

Thermal Energy (E) = 0.37 kW(t)-hr/lb

Process	Electric energy kW(e)-hr/lb	Equivalent G value	Equivalent thermal energy,* kW(t)/lb	Overall thermal efficiency, %
Electric discharge	3.75	6.8	12.5	3.0
	5.0	5.1	16.7	2.2
Radiation		10	7.2	5.3
		15	4.8	7.7

*The power-conversion efficiency is 30% for the electric-discharge process and the energy-deposition efficiency is 35% for the radiation process.

lent to a G value (molecules of O_3 per 100 eV of energy deposited in the reactant, O_2) ranging from 6.8 to 5.1, while G values ranging from 10 to 15 can be obtained using high-energy radiation.

Furthermore, the quantity of energy for synthesizing ozone by high-energy radiation is considerably lower than that for the electric-discharge process. The electric discharge is produced by converting thermal energy in a power plant to electrical energy and subsequently transforming to high voltage, so that the overall thermal efficiency of the process from fuel to chemical is no more than about 2.2 to 3.0%. In the high-energy-radiation process, the radiation is produced directly from nuclear fuel and is converted to chemical energy by direct energy deposition in the oxygen. Not all the high-energy radiation is available to the reactant, some being absorbed in the radiation source and some in the container, but as much as 35% can be transferred to the oxygen. The overall thermal efficiency is then 5.3 to 7.7%, or two to three times that of the electric-discharge process.

HISTORY

Since the early work of Lind,[6] the radiation chemistry of ozone has been studied in experiments performed mainly in static systems, at low pressures and low temperatures. In liquid oxygen in glass apparatus, an initial $G(O_3)$ value of 6 for ozone formation and concentrations up to 6 wt.% are reported by Wall and Brown.[7] In gaseous oxygen at −78°C, Kircher[8] reported a $G(O_3)$ value* of 9 using a steel system, and Johnson and Warman,[9] using 1 atm pressure, reported an initial $G(O_3)$ value of 12.8 with a glass system. In experiments by Sears and Sutherland using glass capsules containing the gas phase at 1 atm in a ^{60}Co gamma field, initial $G(O_3)$ values up to 10.5 were found,[10] while with fission fragments in an in-pile experiment, at a temperature between room temperature and −78°C, a minimum $G(O_3)$ value of 6.9 and concentrations up to 0.5% were obtained.[11]

Investigations have been made in flow systems with air at room temperature at 1 atm pressure to investigate the hazards of ozone formation associated with radiation processes for sterilization of medical supplies and food preservation.[12,13] In a closed glass system, O_3 concentrations up to 18.5 ppM were measured, and a $G(O_3)$ value up to 4.0 can be estimated from the work of Kertesz and Parsons.[13] A study by Shah and Maxie,[14] using air at atmospheric pressure and gamma radiation in a flowing system constructed of glass, indicated a $G(O_3)$ value of 11 with an O_3 concentration of 1.7 ppM. Flow experiments in a high-pressure steel system in a ^{60}Co gamma field at 68 atm and higher with air at room temperature indicated[15] a $G(O_3)$ value of 4.7. Recent experiments[16] with oxygen at pressures up to 200 atm and a temperature of −60°C in a stainless-steel system in a radiation field gave a G value of 10.5.

A G value of 13.8 was determined in pulse radiolysis experiments[17] with high-energy electrons in oxygen at 1 atm and high dose rates.

OZONE SYNTHESIS TESTS

Radiation synthesis of ozone from oxygen was studied in a loop[18] installed in the Brookhaven graphite research reactor. The loop, a doubly contained recirculating-gas system inserted into the lower section of the reactor core, is designed to utilize reactor radiation (neutron and gamma) either directly from the reactor or from the fission-fragment energy of ^{235}U, which is in a foil fuel form especially fabricated[19,20] to efficiently impart this energy to the irradiation system. The fuel is inserted and removed by a drive mechanism.

The results of preliminary tests indicated that the ozone yield is favored by low temperatures because of the thermal instability of ozone, and high yields were obtained between −20 and 0°C. Yields increased with increasing pressure up to 60 atm, the highest tested. The steady-state ozone concentration was almost a direct function of the radiation intensity, but the type of radiation did not affect the yield. Thus the gamma and beta energy from radioisotopic sources (e.g., ^{60}Co, ^{137}Cs, or ^{90}Sr), the gamma and beta radiation from a nuclear reactor, the radiation from uranium fission fragments, and the radiation from electron accelerators should give comparable ozone yields with comparable radiation intensities. The addition of nitrogen to the oxygen feed greatly increased the ozone yield, possibly by forming NO_2, which acts as an electron scavenger to inhibit ozone decomposition, or by coating the stainless-steel walls and thus preventing ozone decomposition on the walls.

In a typical run using reactor radiation (Fig. 1), an ozone concentration of 500 ppM was obtained despite the relatively low radiation dose rate (2.4 Mrads/hr). The integral G value, with 15.6% N_2, was calculated to be about 8. Subsequent work has shown that G values can go as high as 12. The maximum N_2 concentration

*The G value is the molecules of product formed per 100 eV of energy deposited.

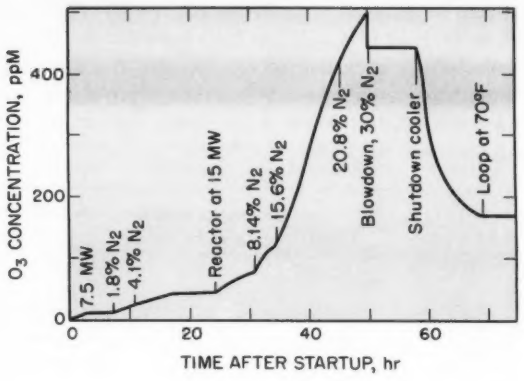

Fig. 1 Data from a typical O_2-O_3 run in the Brookhaven graphite research reactor in-pile loop. Pressure = 800 psig; initial temperature = −30°F; nitrogen added at times indicated. Run stopped at reactor shutdown (about 75 hr).

investigated was 30%, but the optimum concentration has not been determined.

Ozone can be produced continuously in a steel processing system after the surface has been conditioned.

RADIATION PROCESS FOR OZONE PRODUCTION

For economic reasons (see below), two different radiation processes for ozone production were designed: the chemonuclear-reactor process for large capacities and the radioisotopic-source process for small capacities.

Chemonuclear Process

The chemonuclear process for ozone synthesis (Fig. 2) features three basic systems: (1) a system for

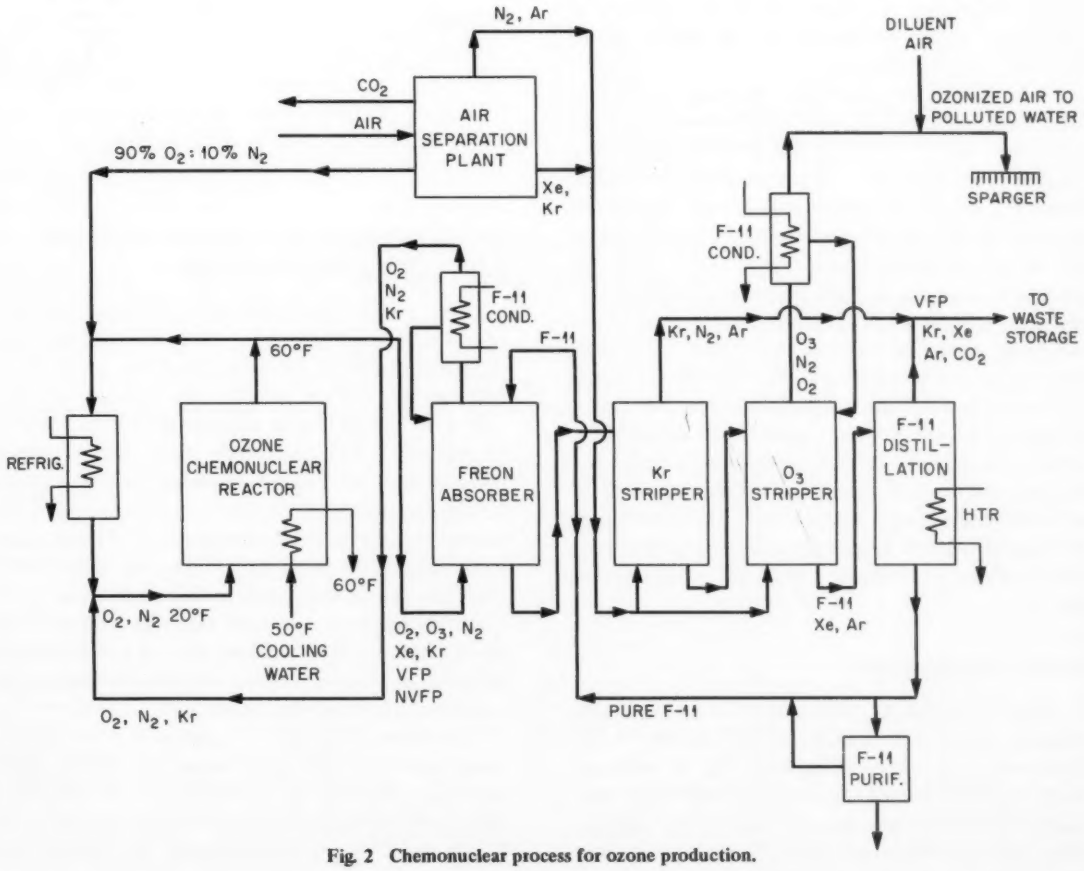

Fig. 2 Chemonuclear process for ozone production.

XUM

recovering oxygen from the air, which is of standard design; (2) a fission-fragment chemonuclear reactor to convert recycled oxygen to ozone; and (3) an absorption–stripping system to recover and decontaminate the ozone.

The chemonuclear reactor would use a fuel of U–Al alloy (20 wt.% U, 93.5% enriched) deposited in a 0.25-mil-thick layer on each side of a 10-mil-thick aluminum plate. This fuel would release at least 20% of the fission-fragment energy into the gas stream, which would be at an average temperature of 30°F and a pressure of 800 psig. The reactor core would be a sphere about 8 ft in diameter and would contain a minimum of 150 kg of ^{235}U.

Feasibility calculations[20,21] have shown that critical assemblies for such a system can be achieved in which most of the heat is removed by a light-water moderator–coolant and the remainder by the process gas, which is then externally cooled. Since the fuel is in direct contact with the reactants, the product ozone would contain uranium fission products, but the nonvolatile fission fragments—cesium, barium, and strontium—would be removed by deposition on reactor surfaces and filtration.

A three-column absorption–stripping system is being considered for removing volatile fission products from the ozone. This system is potentially very efficient and economic. The first column contains Freon 11 or 12, in which ozone and the fission products xenon and krypton are soluble and in which the ozone product is very stable.[22,23] A Freon absorption–stripping system for recovering the noble-gas fission products from the off-gas of a nuclear-fuel-processing plant has been demonstrated on a pilot-plant scale.[24] The krypton would be stripped in the second column and the ozone in the third, the xenon remaining in the Freon. Argon that entered the system with the oxygen feed would mostly follow the krypton, while small amounts of $^{14}CO_2$ present [^{14}C is formed by the (n,p) reaction on ^{15}N] would follow the xenon. Iodine and any nonvolatile fission fragments carried over would remain in the absorption column.

Isotopic Radiation Source

When a separated radioisotope is used as the radiation source, the system is very similar to the electric-discharge ozonizer system (Fig. 3), with an isotopic irradiator replacing the electric-discharge unit. Oxygen separated from the air is sent to the irradiator, where part is converted to ozone. The ozonized oxygen stream then goes to a tower contactor, where the water to be purified is found. The gases from the purifier are collected for recycling of the oxygen, the CO_2 and water being scrubbed out before the oxygen is returned to the irradiator. There is no direct contact of the product with the fuel.

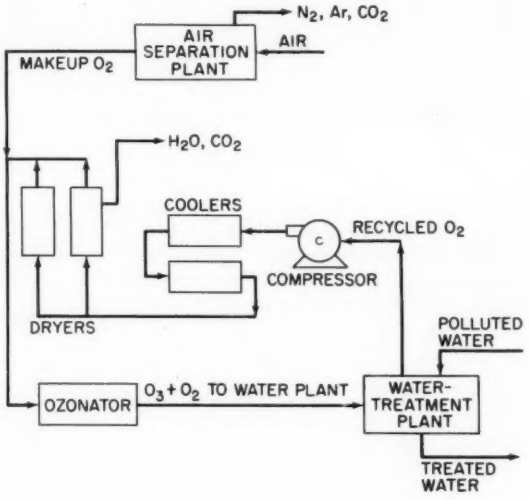

Fig. 3 Ozone process for water treatment.

ECONOMICS OF RADIATION PROCESS FOR OZONE PRODUCTION

The energy requirements for ozone-production plants with capacities of 1 and 400 tons/day were estimated (Table 2). An ozone plant with a capacity of 400 tons/day could supply the ozone to treat 1.28×10^9 gal of sewage effluent per day at a level of 75 ppM O_3. The data show that fission-fragment chemonuclear reactors are indicated for high-capacity tertiary-treatment plants since reactor power levels become economic in the hundreds of thermal megawatts region, the sizes needed for the higher ozone capacities. Single-purpose (n,γ) reactors containing clad fuel elements have a low efficiency and are too large for the smaller capacities, but there is a possibility of using the by-product neutron and gamma energy from a steam- or power-producing reactor.[25]

An ozone plant with a capacity of 1 ton per day could treat 1.2×10^8 gal of water per day for drinking-water purposes or 3.2×10^6 gal of secondary sewage effluent per day. The isotopic sources ^{60}Co, ^{137}Cs, and ^{90}Sr are indicated only for relatively low

Table 2 Possible Radiation Processes for Ozone Production

G = 15; Energy Equivalent = 1.7 kW(t)-hr/lb O_3

Radiation source	Energy-deposition efficiency, %	Total energy, kW(t)-hr/lb O_3	Size of reactor or source
400 tons of O_3 per day			
Fission	35	4.8	560 MW(t)
fragments	10	16.9	
1 ton of O_3 per day			
Fission	35	4.8	0.4 MW(t)
fragments	10	16.9	1.4 MW(t)
Reactor,	3	56.3	4.7 MW(t)
(n,γ)	0.5	338.0	28.2 MW(t)
^{60}Co gamma	50	3.4	19.3 MCi
^{137}Cs gamma	50	3.4	86.0 MCi
^{90}Sr beta	50	3.4	42.5 MCi

volumes, e.g., secondary sewage effluent of less than 10^7 gal/day. Even at the lowest cost of $0.10 per curie, ^{60}Co would require an investment of almost 2×10^6 for a 1 ton/day plant, while electric-discharge ozonizers would cost less than 1×10^6. Furthermore, the availability of ^{60}Co would be a problem. A possibility exists of obtaining larger quantities of ^{137}Cs from fission-product wastes as the nuclear power industry expands in the United States in the next decade, but the cost of recovery and encapsulation, and taking credit for long-term storage, would control its usage for this purpose.

Chemonuclear Vs. Electric-Discharge Process

Estimates of capital investment and ozone-production costs, as a function of plant capacity from 40 to 400 tons of ozone per day (Tables 3 and 4 and Fig. 4), show that with a G value of 10 and an energy-deposition efficiency of 20%, the chemonuclear and electric-discharge ozonizer costs are the same for a 220 ton/day plant. At either higher G values or higher energy-deposition efficiencies, the chemonuclear plant is more economical than the electric discharge at almost all capacities considered. For the chemonuclear process, a G value of 10 and an efficiency of 20% are reasonably attainable, and a G value of 15 and an efficiency of 35% are ultimately attainable. For the electric-discharge process, an energy consumption of 5 kW(e)-hr/lb was used as probable, with an ultimate value of 3.75 kW(e)-hr/lb. An additional 1.25 kW(e)-hr/lb was added to account for powering auxiliary equipment, such as the oxygen-plant recycle compressors and gas coolers and dryers.

Table 3 Cost of Chemonuclear-Plant Ozone

Plant Capacity: 400 tons/day

	G = 10		G = 15	
	E = 0.20, reactor power = 424 MW(t)	E = 0.35, reactor power = 242 MW(t)	E = 0.20, reactor power = 284 MW(t)	E = 0.35, reactor power = 160 MW(t)
	Investment Costs, $10^6			
Oxygen plant	3.50	3.50	3.50	3.50
Refrigeration	4.30	2.50	3.00	1.70
Reactor	17.40	17.40	17.40	17.40
Freon plant	7.00	7.00	7.00	7.00
Compressors	1.60	0.83	0.83	0.70
	33.80	31.23	31.73	30.30
	Operating Costs, cents/lb O_3			
Depreciation at 7.3%	0.95	0.91	0.91	0.89
Nuclear fuel	1.30	0.80	0.90	0.50
Labor	0.08	0.08	0.08	0.08
Maintenance	0.08	0.08	0.08	0.08
Power	1.14	0.72	0.81	0.53
	3.55	2.59	2.78	2.08

Note: E = energy-deposition efficiency.

Table 4 Energy Requirements for Chemonuclear and Electric-Discharge Ozone

	Energy requirement, kW-hr/lb O_3	
	G = 10, E = 0.20 reactor thermal power of 12.80*	G = 15, E = 0.35 reactor thermal power of 4.85*
Chemonuclear plant		
O_2 plant, electrical	0.21	0.21
Refrigeration, electrical	0.95	0.33
Compressors, electrical	1.05	0.39
	2.21	0.93
Electric-discharge plant		
O_2 plant	0.21	0.21
Electric ozonizers	3.75	5.00
Auxiliaries	1.04	1.04
	5.00	6.25

*Thermal-energy output of nuclear reactor required for ozone production is in kW(t)-hr per pound of ozone.

Because of the high electrical consumption required for conventional ozonizers, power costs were estimated at two values—5 and 6 mills/kW-hr. The bases for the chemonuclear plant estimates were:

1. 20% of the reactor heat is removed by recycle-gas refrigeration, and the remainder by cooling water.

2. Power cost of 5 mills/kW-hr.

3. Gas-refrigerant cost of $200 per ton.

4. Annual fixed-charge rate on capital investment is 7.3%, based primarily on municipal water-treatment plants (for chemical oxidation plants, the rate is nearer 25%; see below).

5. Labor for the 200 and 400 ton/day plants is six men per shift, and for the 40 and 100 ton/day plants, five men per shift each plant, with three shifts at $10,000 per man-year.

6. Maintenance cost equal to labor cost for the 400 ton/day plant and scaled for the smaller plants by a 0.7 power factor of plant capacity.

7. Compressor, refrigeration, and oxygen-plant costs obtained from commercial vendors.

8. Reactor costs obtained from estimates by Burns and Roe[26] and from nuclear-reactor price books of General Electric and Westinghouse.

9. Freon system estimated at twice the cost of the oxygen plant because of additional columns and shielding.

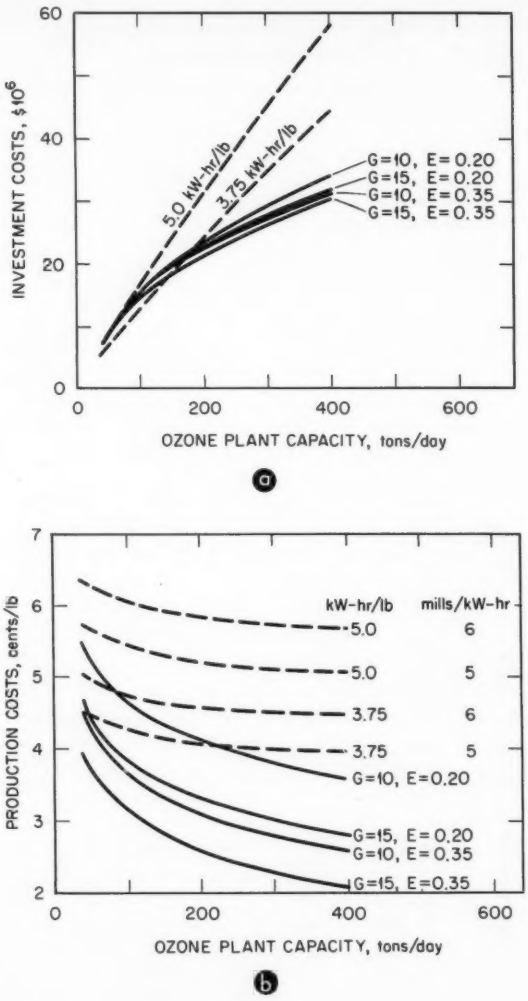

Fig. 4 (*a*) Investment and (*b*) production costs for ozone-production plants. ---, electric discharge; ——, chemonuclear. E = fission-fragment energy-deposition efficiency.

For the conventional electric-discharge ozonizer system, the bases were:

1. Cost of ozonizer and recycle equipment obtained from Welsbach Corporation Bulletin 201 (Ref. 5) and extrapolated to the larger capacity by a 0.9 power factor of capacity.

2. Fixed-charge rate on capital of 7.3%.

3. Labor cost half that of chemonuclear.

4. Maintenance cost equal to labor cost for 400 ton/day plant, scaled by a 0.9 power factor.

Table 5 Total Ozone-Production Costs at 25% Fixed-Charge Rate

	Costs, cents/lb O_3			
	40 tons/day		400 tons/day	
	G = 10, E = 0.20	G = 10, E = 0.35	G = 15, E = 0.20	G = 15, E = 0.35
Chemonuclear plant				
Depreciation at 25%	8.08	7.98	3.12	3.05
Nuclear fuel cycle	1.30	0.80	0.90	0.50
Labor	0.50	0.50	0.08	0.08
Maintenance	0.16	0.16	0.08	0.08
Power	1.14	0.72	0.81	0.53
	11.18	10.16	4.99	4.24
	3.75 kW-hr/lb	5.00 kW-hr/lb	3.75 kW-hr/lb	5.00 kW-hr/lb
Electric-discharge ozonizer plant				
Depreciation at 25%	5.90	7.52	4.66	6.10
Power cost				
At 5 mills/kW-hr	2.50	3.12	2.50	3.12
At 6 mills/kW-hr	3.00	3.75	3.00	3.75
Labor	0.25	0.25	0.04	0.04
Maintenance	0.05	0.05	0.04	0.04
Total				
At 5 mills/kW-hr	8.70	10.94	7.24	9.30
At 6 mills/kW-hr	9.20	11.57	7.74	9.93

Table 5 shows production costs for the chemonuclear and the electric-discharge methods at a fixed-charge rate of 25%, which is an annual rate commonly used in the chemical industry. The table points out the costs for the small and large plant capacities of 40 and 400 tons/day.

The results of the cost analysis indicate that, under optimum yield conditions, ozone can be produced in the chemonuclear system for 1 to 2 cents per pound less than in conventional electric-discharge ozonizer plants. For large capacities, up to 400 tons/day, the optimum chemonuclear cost is half that of the electric-discharge cost, and, under almost any conditions lower than optimum, the chemonuclear process is less costly. The above conclusions hold for water-treatment plants where depreciation rates are low (7.3%). For the higher depreciation rates (25%) used in chemical plants, there is a decided cost advantage—as much as 2 to 6 cents per pound where larger capacities are needed.

An additional factor in favor of the chemonuclear system is that it is inherently a large-volume high-capacity system, while the electric-discharge ozonizers reach optimum in a single unit at relatively low capacities and must be duplicated many times for high capacity, introducing severe maintenance problems.

The treatment cost (Table 6) for 1.28×10^8 gal of 20-ppM-COD water per day with the electric-discharge plant would be 2.8 cents per 10^3 gal and with the chemonuclear plant, 0.3 cent per 10^3 gal less. However, at 4000 tons/day the cost for treating 1.28×10^9 gal/day is decreased from 2.5 cents to 1.25 cents per 10^3 gal. If water with 40 ppM COD is treated, the saving with the chemonuclear process is even more substantial, amounting to 2.5 cents per 10^3 gal.

Projected estimates[27] for activated-charcoal treatment indicate costs of 5 to 6 cents per 10^3 gal for capacities of the order of 10^8 gal/day. Thus substantial savings in high-capacity water-treatment plants can result from the use of the chemonuclear ozone process. It is understood that a number of plants in the country will be processing sewage in volumes of from 10^8 to 10^9 gal/day range within the next decade. The incentive is thus large for further development of a chemonuclear process. (MG)

Table 6 Water-Treatment Costs

COD reduction, ppM	O_3 capacity, tons/day	Sewage vol. treated, 10^9 gal/day	O_3 cost, cents/lb	Water-treatment cost, cents/10^3 gal
		Electric-Discharge Ozonizer Process		
20	40	0.128	4.5	2.8
20	400	1.28	4.0	2.5
40	400	0.64	4.0	5.0
		Chemonuclear Process		
20	40	0.128	4.0	2.5
20	400	1.28	2.0	1.25
40	400	0.64	2.0	2.5

XUM

REFERENCES

References marked with an asterisk have not been verified by the editors; for further explanation, see the Foreword.

1. Ozone Chemistry and Technology, *Advan. Chem. Ser.*, No. 21 (1959), American Chemical Society, Washington, D. C.
2. D. C. O'Donovan, Treatment with Ozone, *J. Amer. Water Works Ass.*, **57**: 1167-1194 (1965).
3. A. Halfon, D. T. A. Huibers, and R. McNabney, paper presented at American Chemical Society Meeting, Atlantic City, Sept. 8–13, 1968 (Abstr. Watr-031 in *Abstracts of Papers*).
4. T. C. Manley and S. J. Niegowski, Ozone, in *Encyclopedia of Chemical Technology*, Vol. 14, 410-432, 2nd ed., Interscience Publishers, New York, 1967.
5. *Welsbach Corporation, Philadelphia, Pa., Technical Bulletin No. 201, 1968.
6. S. C. Lind, *Radiation Chemistry of Gases*, pp. 83-86, Reinhold Publishing Corporation, New York, 1961.
7. D. W. Brown and L. A. Wall, Gamma Irradiation of Liquid and Solid Oxygen, *J. Phys. Chem.*, **65**: 915-919 (1961).
8. J. F. Kircher, J. S. McNulty, J. L. McFarling, and A. Levy, Irradiation of Gaseous and Liquid Oxygen, *Radiat. Res.*, **13**: 452-465 (1960).
9. G. R. A. Johnson and J. M. Warman, Formation of Ozone from Oxygen by the Action of Ionizing Radiations, *Discuss. Faraday Soc.*, **37**: 87-95 (1964).
10. J. T. Sears and J. W. Sutherland, Radiolytic Formation and Decomposition of Ozone, *J. Phys. Chem.*, **72**: 1166-1171 (1968).
11. J. T. Sears and J. W. Sutherland, Estimates of Ozone Yields in Proposed Chemonuclear Reactors, *Nucl. Appl.*, **5**: 62-63 (1968).
12. L. N. Less and A. J. Swallow, Estimating the Hazard Due to Radiolytic Products from Air, *Nucleonics*, **22**(9): 58-61 (1964).
13. Z. I. Kertesz and G. F. Parsons, Ozone Formation in Air Exposed to ^{60}Co Gamma Radiation, *Science*, **142**: 1289-1290 (1963).
14. J. Shah and E. C. Maxie, Gamma-Ray Radiosynthesis of Ozone from Air, *Int. J. Appl. Radiat. Isotop.*, **17**: 155-159 (1966).
15. M. Steinberg, O_3 and NO_2 Formation by Irradiation of a N_2–O_2 Gas Mixture in a Flowing System at Elevated Pressures, USAEC Report BNL-50017, Brookhaven National Laboratory, September 1966.
16. R. N. Dietz, J. D. Smith, and M. Steinberg, Effect of Gamma Irradiation on the Oxidation of Nitrogen (Abstr. Nucl-007 in *Abstracts of Papers*) and Low Temperature Gamma Irradiation Synthesis of Ozone (Abstr. Nucl-008 in *Abstracts of Papers*), papers presented at American Chemical Society Meeting, New York, Sept. 7–12, 1969.
17. J. A. Ghormley, C. J. Hochanadel, and J. W. Boyle, Yield of Ozone in the Pulse Radiolysis of Gaseous Oxygen at a Very High Dose Rate: Use of This System as a Dosimeter, *J. Chem. Phys.*, **50**: 419-423 (1969).
18. M. Steinberg, W. Tucker, C. Waide, M. Beller, P. Bezler, and B. Manowitz, Safety Review of the Brookhaven Chemonuclear In-Pile Research Loop, USAEC Report BNL-13001, Brookhaven National Laboratory, October 1968.
19. M. Steinberg, W. Tucker, and G. Farber, Fission-Fragment Recoil Energy Source and Chemonuclear Fuel Element, *Trans. Amer. Nuclear Soc.*, **10**: 44-45 (1967).
20. M. Steinberg, Fission Recoil Synthesis, *Chem. Eng. Progr.*, **62**(9): 105-116 (1966).
21. M. Beller, D. Goellner, and M. Steinberg, Design and Economic Study of a Gas-Phase Chemonuclear Ozone Process, *Nucl. Appl.*, **1**: 322-326 (1965).
22. F. Mahieux, Preservation of Ozone, French Patent No. 1,246,273 (Oct. 6, 1959) (*Chem. Abstr.*, **56**: 1150*b*).
23. M. Steinberg, The Recovery of Fission Product Xenon and Krypton by Absorption Processes, USAEC Report BNL-542, Brookhaven National Laboratory, 1959.
24. J. R. Merriman, J. H. Pashley, K. E. Habiger, M. J. Stephenson, and L. W. Anderson, Concentration and Collection of Krypton and Xenon by Selective Absorption in Fluorocarbon Solvents, in *Treatment of Airborne Radioactive Wastes*, Symposium Proceedings, New York, 1968, pp. 303-313, International Atomic Energy Agency, Vienna 1968 (STI/PUB/195).
25. M. Beller and M. Steinberg, The Possibility of Chemonuclear Reactors Producing Ozone for Water Treatment, *Nucl. Appl.*, **2**: 237-238 (1966).
26. Burns and Roe Engineering Company, Oradell, N. J., personal communication, March 1969.
27. *R. Smith, Cost and Performance Estimates for Waste Water Treating Processes To Be Used Downstream of Secondary Treatment, Federal Water Pollution Control Administration, Cincinnati, Ohio, 1969.

XUM

Sewage and Waste-Water Processing with Isotopic Radiation: Survey of the Literature

By Martha Gerrard

Supplementary Keywords: *water and waste treatment; sterilization; source, gamma; economics; accelerator; X-ray machine; pollution; industry, utility; industry, mining; industry, chemical;* ^{60}Co; ^{90}Sr; ^{137}Cs; ^{144}Ce.

Abstract: *The literature (~65 references) on the use of ionizing radiation to process domestic and industrial liquid waste is reviewed.*

Ionizing radiation has the ability to neutralize charges on colloidal or suspended particles, to destroy microorganisms, and to oxidize or otherwise decompose or change the structure of organic and inorganic compounds. Because of these properties, it is of interest to processors of domestic and industrial wastes, as suggested by Ballantine et al.,[1] for improving sludge settleability, for disinfection, and for destruction of organic and inorganic compounds. The effluent from a plant that treats waste with radiation contains no induced activity.

A pioneer in sewage-irradiation work was Dunn,[2] at the Massachusetts Institute of Technology, who first studied the gamma-radiation processing of foods, which have a low bacterial population. He subsequently turned his attention to sewage, with its high bacterial population, and was able to sterilize sewage samples with <1 Mrad* of either ^{60}Co gamma or accelerator (Van de Graaff) radiation. A few years later, the Illinois Institute of Technology Research Institute[3] compared 10 sewage-processing methods—several types of ionizing irradiation, ultraviolet irradiation, chemical treatment, and several heat treatments. Irradiation with an isotopic source was one of several recommended for further study. For a 5 gal/min plant, with 4 Mreps necessary for sewage sterilization, this technique was concluded to be feasible if isotopic source costs should be decreased, isotope availability increased, and some means found for increasing microorganism sensitivity. Cost estimates were based on ^{60}Co at $5 per curie.† Since this early work, the direct irradiation technique has been studied at a number of sites for both sewage and industrial waste-water processing. The basic principles of the process have been described by Ballantine.[1]

The published literature on large-scale direct irradiation, as indicated by a search of *Nuclear Science Abstracts* and *Chemical Abstracts*, is reviewed here. (The indirect use of isotopic irradiation in sewage processing, i.e., radiation conversion of oxygen to ozone for use in sewage processing, is not included; but see the article by Steinberg and Beller in this issue of *Isotopes and Radiation Technology*.) Earlier reviews on one or more phases of sewage- and waste-irradiation work are listed in Ref. 4.

SEDIMENTATION OF SUSPENSIONS

Sludge

Sludge, the particulate mass formed by gravity separation of sewage, has a solids concentration of about 2.5 lb per 100 lb of sludge. In ordinary practice, sludge is anaerobically digested 15 to 30 days, as a result of which the solids content is decreased to 0.5 to 1.0 lb/100 lb. This digest is treated with chemicals, filtered, and buried or sometimes is further treated and used as fertilizer. Digestion may be hastened by activation of the raw sludge, i.e., by adding to the raw sludge a sludge that is already biologically active.

Etzel et al.,[5] using sludge from a Chicago treatment plant, increased the rate of settleability of waste-activated sludge two- to three-fold by gamma (^{60}Co) irradiation with a total dose of 2×10^5 rads. Irradiation of digested sludge did not increase its rate of settling but did increase its rate of filtration.

In work[6] started in 1960 at Unidynamics, under a Division of Isotopes Development contract, it was also found that irradiation increased sewage sedimentation. A dose of 1270 rads, at a dose rate of 10.4 R/min from a 2-kCi ^{60}Co source, increased sedimentation 6%, and 13,200 rads at 296 R/min increased it 8%. The sedimentation achieved at a given total dose depended on the dose rate (Fig. 1). For example, at a rate of 256 R/min, all suspensions given a total dose of

*1 rad ≈ to 1 R absorbed; 1 rep ≈ 0.93 R absorbed. In this article the units are quoted as used in the originals.

†The current ORNL catalog quotes $0.50 per curie in 10^6-Ci lots for material with a specific activity of 30 Ci/g, which is sufficiently high for a sewage-processing source.

XUM

35 krads or more showed increased sedimentation. However, at higher dose rates—507 and 643 R/min—there was either no change or a decrease in sedimentation for all total doses, while at 850 and 1340 R/min, sedimentation again increased. Sedimentation was maximum—19%—with 280 krads at a rate of 850 R/min.

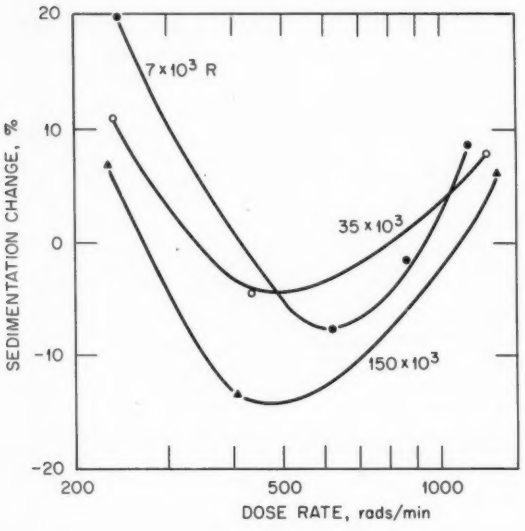

Fig. 1 Dependence of sedimentation due to gamma (^{60}Co) irradiation on total dose and dose rate.

In Canadian studies,[7] the precipitation of carbonaceous matter from raw sewage by lime was somewhat enhanced by irradiation. The filtrate was sterile owing to removal of bacteria in the lime slurry.

Other Suspensions

Unidynamics also investigated[8] the effect of gamma (^{60}Co) radiation on colloidal suspensions of industrially important water-soluble polymers—poly(vinyl alcohols), gums, pectins, polymeric ethers, methyl cellulose, and others; on nonionic surfactants; and on natural oil emulsions. They found that irradiation increased the sedimentation rate of the colloidal suspensions, thus indicating the feasibility of radiation processing of certain industrial waste waters. The surfactants were functionally changed, but the oil emulsions were stabilized under certain conditions.

At the Georgia Institute of Technology Experiment Station, the effects of both X and gamma irradiation on suspensions of stannic oxide, aluminum oxide, ferric oxide, bacteria, and natural river-water components were investigated.[9] Of all these suspensions, only bacteria were precipitated, and a high radiation dose with a low *p*H was necessary for that. Smith and Ledbetter[10] found that gamma (^{60}Co) irradiation (up to 300 R) of a kaolinite suspension did not change the amount of alum needed to coagulate the suspension, while Grune[11] found that 20 kR of X radiation had no measurable effect on Al_2O_3 sols, but that 120 kR induced some coagulation in Fe_2O_3 sols.

Cornstarch sols were found by Tyler, Munno, and Cadman[12] to be progressively degraded by gamma (^{60}Co) irradiation with doses greater than 10^4 rads. There was no enhancement of sedimentation with 10^2 to 10^4 rads, and from 10^4 to 10^6 rads there was some decrease in the sedimentation.

Russian workers[13] irradiated mineral sludges (galenite, pyrite, quartz, calcite, albite, fluorite, and bosite) with the beta radiation from ^{90}Sr–^{90}Y and ^{144}Ce–^{144}Pr sources for 30 min to 6 hr and found a decrease in the rate of precipitation 1 hr after exposure. The radiation-coagulated specimens had larger and more compact particles, were more dense, and contained less water and therefore filtered more rapidly.

DISINFECTION

Pathogenic microorganisms in sewage must be destroyed or essentially removed before the treatment-plant effluent can be released to the environment. In early studies using gamma (^{60}Co) radiation, Ridenour and Armbruster,[14] using pure cultures of 10 typical organisms found in domestic sewage, obtained 99% or more destruction with 10^5 reps, with coliform the most susceptible and *Streptococcus faecalis* and *Bacillus subtilis* the most resistant. Higher radiation doses were necessary to produce the same effect in sewage, probably because of the action of organic matter and clumping of organisms. In the primary plant effluent, 10^5 reps produced 99% kill, and, in the final effluent, half this dose was required for a similar decrease. The *p*H had no effect in the range 5 to 8.

Lowe et al.[15] found that in distilled water the gamma (^{60}Co) irradiation necessary for complete kill ranged from 7.4×10^4 rads for *Escherichia coli* phage T-3 to 2.0×10^6 rads for *B. subtilis. Mycobacterium smegmatis, Micrococcus pyrogenes* var. aureus, and *E. coli* fell between these limits. In autoclaved sewage cultures, 5.0×10^6 rads was necessary for 100% kill, and in air-dried sludge, 2.0×10^6 rads. Touhill et al.[16]

XUM

studied five principal types of microorganisms often found in secondary waste-water effluent and reported 99.995% kill at a dose of 2.5×10^5 rads. The enterococci and *E. coli* phages were approximately 100% killed and spores, 92%.

In Italian work,[17] 100% kill of the total microbe population in a sedimented sewage was obtained with 3.96×10^6 rads, although the coliforms were 100% killed with 1.5×10^5 rads.

In Romanian laboratory-scale studies[18] of gamma (^{60}Co) irradiation of waste water, 88.89 to 99.51% of the major germs were destroyed by 3×10^5 R. In these studies, test tubes containing the waste water were rotated around the 25-Ci source. Based on the results of this work, possible designs for a practical installation were studied.

Ryabchenko[19a] sterilized water containing *E. coli, Salmonella paratyphi, Salm. typhi,* and *Shigella sonnei* with 40 to 50 kR of gamma (^{60}Co) radiation. Since the *E. coli* required the highest dose, he concluded that the *E. coli* content retains its significance as a sanitary index of irradiated water. However, he subsequently reversed[19b] his opinion upon finding *E. coli* less resistant than enteroviruses and the poliomyelitis virus.

An ionizing radiation dose of 10^5 R is reported[20] to inhibit growth of *Endamoeba histolytica.*

The bacterial count of water that had been used in washing grain was sharply decreased by gamma (^{60}Co) irradiation, but pretreatment, e.g., with a chemical or ultrasound, was necessary for complete sterilization.[21]

Fowler[22] expressed the opinion that sewage treatment may alter the food supply available to microorganisms rather than the microbial population per se.

A patent[23] has been issued for a method of killing bacteria in underground fluids (oil and water) by placing a source made of radioactive waste in the path of such fluids.

Irradiation enhances the sterilizing effect of chlorine. In a Russian experiment,[24] waste was sterilized with 1 mg of chlorine per liter plus a radiation dose of 2.5×10^4 R. This was one-tenth the amount of chlorine required in the absence of radiation for the waste samples under study. Similar results were obtained in Florida in a test plant.[25]

DESTRUCTION OF ORGANIC AND INORGANIC COMPOUNDS

Organic

The Unidynamics study[6] of sewage showed that gamma irradiation could eliminate detergent foaming by degradation of the detergent molecule. Doses of less than 10^4 rads destroyed 50% of the alkylbenzene sulfonate (ABS) present in a solution of 14 ppM concentration, and 9×10^4 rads at a rate of 10.4 R/min or 5×10^4 rads at a rate of 22.6 R/min destroyed 99% of the detergent (Fig. 2). Even 1300 rads, which resulted in a 6 to 8% increase in sedimentation, produced a 25% decrease in ABS. In a study of the effects of various parameters, Fleischman and Price[26] found 98% of the ABS present destroyed by 5.4×10^4 rads (^{60}Co).

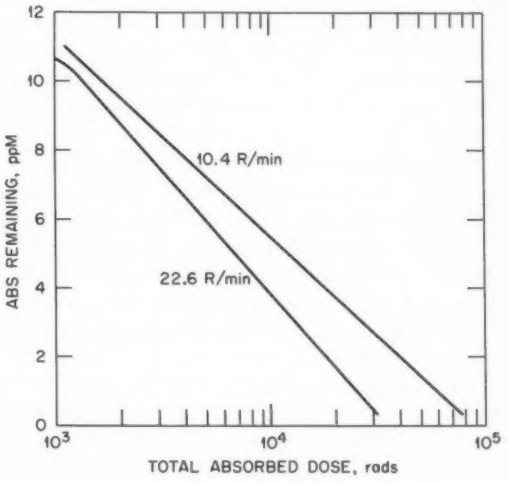

Fig. 2 Dependence of degradation of ABS on total absorbed dose and dose rate.

Russian laboratory studies[27] have also shown decomposition of several ionogenic sulfonates to a biodegradable product: Nekal, a mixture of mono-, di-, and tributylnaphthalene sulfonates; Sulfonol, a tetramer of propylene and the kerosene fraction of petroleum; and STEK, a sulfonate of paraffin and aromatic hydrocarbons of unknown molecular weight. Degradation increased with increasing sulfonate concentration in the range 50 to 500 mg/liter, with passage of oxygen through the solutions, with decrease in *p*H to 1–2, and with a decrease in dose rate from 200 to 6 R/sec. Nekal solutions of 100 mg/liter concentration were 100% destroyed by 0.3 to 0.5 Mrad. There was no difference in the radiolysis of the three materials although their biological degradation differs considerably. Radiolysis of two nonionogenic compounds did not differ essentially from that of the ionogens.

XUM

In a Japanese study,[28] 10 kR of ^{60}Co gamma irradiation of solutions containing ppM quantities of ABS, phenol, parathion, and phenylmercuric acetate destroyed more than 80% of the contamination. Yields were poorer with samples containing higher concentrations. Mice showed no abnormalities when fed the filtered product.

Touhill et al.[16] found phenol in concentrations of 1000 mg/liter to be 48% destroyed in 5 hr and 99% in 20 hr with a ^{60}Co gamma dose rate of 1.74×10^5 R/hr. When oxygen was added, 75% of the phenol in 100 mg/liter solutions was destroyed in 1 hr. Dodecane and normal paraffin hydrocarbons, simulating constituents of petroleum-related wastes, were degraded to a limited extent by gamma irradiation. Alexander[29] found the biodegradability of the primary effluent of a sewage plant to be decreased by ^{60}Co gamma irradiation.

Condren and Etzel[30] observed a general trend of increasing chemical-oxygen-demand (COD) removal from simulated waste water with increasing dose of ^{60}Co gamma radiation, the reaction being increased by addition of oxygen—e.g., through aeration or by introducing H_2O_2 (Fig. 3). They found a >99.99% decrease in coliforms with a radiation dose of 10^6 rads.

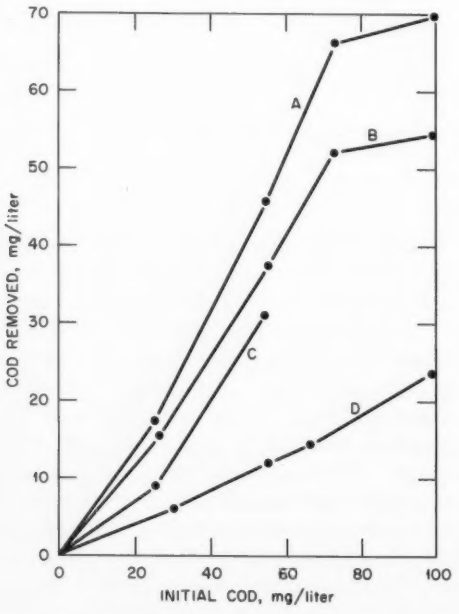

Fig. 3 COD removed by 10^6 R from solutions (*A*) aerated and settled, (*B*) aerated and mixed, (*C*) nonaerated but with 60 mg of H_2O per liter added, and (*D*) nonaerated (control).

Inorganic

Radiation destruction of cyanide, a constituent of certain industrial wastes, and of iron, often found in mine-drainage water, has received attention.

Cyanide. Touhill et al.[16] were able to completely degrade cyanide in concentrations of 0.5 to 45 mg/liter, those normally found in Chicago wastes. One hour was required at a dose rate of 1.74×10^4 R/hr, and neither change in temperature in the range 26 to 75°C nor change in oxygen concentration had any effect. A patent[31] on a procedure for gamma-radiation decomposition of cyanide in waste water has been issued to Western Electric Co.

Italian workers[32] gamma (^{137}Cs)-irradiated cyanide solutions of 10 to 75 ppM concentration at rates of 0.717×10^6 R/hr. At *p*H 9, with a total dose of 0.21 Mrad, cyanide in the 10-ppM solutions was totally destroyed. A 10-fold increase in total dose destroyed 98% of the cyanide in the 75-ppM solutions.

The radiolysis of aqueous cyanide solutions has also been studied in Japan.[33] The decomposition of aqueous KCN solutions by gamma radiation at a dose rate of 1.59×10^5 R/hr for 60 to 80 min was greater the higher the KCN concentration. Ammonia was formed slowly at the beginning of the irradiation but more rapidly as the irradiation continued.

Iron. Work at Brookhaven National Laboratory[34] indicated that iron removal from acid mine-drainage waste with a high iron content (350 ppM) by limestone neutralization could be enhanced by ^{60}Co gamma irradiation. The vigorous mechanical agitation required with the limestone is not necessary when radiation is used. A system for irradiation of this waste was proposed, the radionuclides ^{60}Co and ^{137}Cs being considered and compared with ^{90}Sr.

Sulfates. Solid $FeSO_4$ hydrate and $NiSO_4 \cdot 6H_2O$ were decomposed by gamma (^{60}Co) irradiation, but the sulfates of Na, Mg, Zn, Cu, Cr, and Mn were not.[35]

ECONOMICS

The question of whether the use of radiation for processing of sewage and waste water is economic has not been settled. In 1958, a *Nucleonics*[36] evaluation of radiation for sterilizing sewage stated that building a pipeline 5 miles long to dump sewage in the ocean, far from the shore, was cheaper than using an irradiation process and a short pipeline. A cost of 1.4 cents per gallon for irradiation alone was compared with 0.25 cent per gallon for a combined heat and radiation

XUM

treatment in reaching this conclusion. The article also described a possible treatment method consisting of addition of a nuclide—e.g., ^{10}B—to the sewage and then passage of the sewage stream through a reactor. Neutron capture by the boron would lead to emission of alpha and beta radiations, which would be the effective agents. This system was suggested as being more efficient than direct gamma irradiation because gamma radiation goes right through the stream while the particulate radiation strikes the contaminant particles. An alpha or beta emitter cannot be used as an external source because these rays do not penetrate pipes readily. The possibility of induced radiation, e.g., formation of radioactive sodium, makes the value of such a method problematical.

Also in 1958, Bell[37] suggested that the use of reactor-produced heat to dry fresh activated sludge for disposal might be more practical than direct radiation treatment. He thought, however, that this could possibly induce genetic changes in bacteria.

Of course, the 1958 assumption that raw sewage can be dumped in the ocean even at a distance undoubtedly will not continue to be valid, and in 1967 Narver[38] commented that the combination of irradiation plus a short ocean outfall might become economic in the future. He predicted that radiation processing of drinking water would be practiced earlier than that of sewage, the suspended particulates in sewage being a complication. Ballantine,[1b] however, estimates that, even with optimistic predictions of decreasing both radiation-dose requirements and irradiation costs, radiation treatment will still cost more than conventional methods.

In many cases, however, the economics will be less important than the consideration that no other sewage or waste-water treatment is at all feasible. For example, the irradiation technique could be useful for processing sewage in remote areas, such as the Arctic regions, where disposal facilities are limited.[39] A ^{60}Co irradiation facility,[25] designed by the Florida Institute of Technology, was installed at a fishing camp near Palmdale, Fla., just west of Lake Okeechobee and went on line in March 1970. The effluent is pure enough to use for irrigation, and costs are estimated at $1.477 per 10^3 gal for the 10^4 gal/day plant. A patent on the process has been issued to Energy Systems, Inc. Whittemore et al.,[40] in the same issue of *Nuclear News* where this plant is described, however, emphasized that the applicability of ionizing radiation to treatment of municipal waste water and sewage sludges is likely to be very limited. Although irradiation with doses of 100 to 1000 krads causes significant effects in sewage, similar to those achieved by conventional treatment methods, costs for irradiation are higher. Further, they feel that irradiation may result in partially oxidized compounds of unknown properties which might be a health hazard if discharged. A suggested possible application of irradiation is in modification of industrial waste with high molecular weight, e.g., in decolorizing dyes. Their work was done with spent fuel elements, and they took irradiation costs at 50 cents per 10^3 gal. Other interest in this area is indicated by the conceptual design made for a radiation sewage-processing plant in the Chicago area,[41a] a design of a plant for radiation processing of poultry wastes,[42] and a patent issued to the General Electric Company for sewage treatment using the gamma radiation from cooling fuel elements.[43] Aqueonics, Inc., expects soon to have a radiation-processing system for domestic, restaurant, and laundry waste water in the Sierra Nevada mountains.[44] The Chicago design was based on work[41b] which indicated that the use of 4 million Ci of ^{60}Co or ^{137}Cs should decrease the cost of sludge disposal from $57 to $16 per ton. Mytelka,[45] who worked with nonbiodegradable organic compounds, considers that the overall efficiency of oxidation reactions will have to be increased 2- to 25-fold for radiation processing of sewage to become economic.

Steinberg et al.[34] estimate a cost of $0.05 to $0.25 per 10^3 gal for treating mine-drainage waste with gamma radiation to enhance removal of iron but consider this not to be economic.

Italian workers[32] estimated the cost of destroying cyanide in wastes to be around 1250 lira ($2) per kilogram of CN^-.

REFERENCES

1.*a.* D. S. Ballantine, L. A. Miller, D. F. Bishop, and F. A. Rohrman, The Practicality of Using Atomic Radiation for Wastewater Treatment, *J. Water Pollut. Contr. Fed.*, **41**: 445-458 (1969).

b. D. S. Ballantine, The Potential Role of Radiation in Wastewater Treatment, paper presented at the 62nd Annual Meeting of American Institute of Chemical Engineers, Nov. 16–20, 1969, Washington, D. C. (see also this issue of *Isotop. Radiat. Technol.).*

2. C. G. Dunn, Treatment of Water and Sewage by Ionizing Radiation, *Sewage Ind. Wastes*, **25**: 1277-1281 (1953).

3.*a.* Sterilization of Sewage Containing Pathogenic Microorganisms, Phase I, Revised. Report No. 9 on ARF Project No. 3676, Illinois Institute of Technology, Armour Research Foundation, Apr. 10, 1956 (AD-286430).

b. Ibid., Addendum to Report No. 9, May 5, 1956 (AD-286432).

4.*a.* N. E. Nielson, *Water Treatment Using Gamma Radiation,*

XUM

International Nutronics, Inc., 200 Third St., Los Altos, Calif. 94022, November 1969.

b. D. M. J. Compton, S. J. Black, F. L. Lieurance, and W. L. Whittemore, Application of Ionizing Radiation to the Treatment of Waste Waters and Sewage Sludge, in *Large Radiation Sources for Industrial Processes*, Symposium Proceedings, Munich, 1969, pp. 399-416, International Atomic Energy Agency, Vienna, 1969 (STI/PUB/236) (USAEC Report GA-9632).

5.*a.* J. E. Etzel, G. S. Born, J. Stein, T. J. Helbing, and G. Baney, Sewage Sludge Conditioning and Disinfection by Gamma Irradiation, *Amer. J. Public Health*, **59**: 2067-2076 (1969) (96th Annual Meeting of the American Public Health Association, Detroit, Nov. 11, 1968).

b. Using Gamma Rays for Wastewater Treatment, *Ind. Water Eng.*, **4**: 41 (September 1967).

6.*a.* Investigation of the Effects of Ionizing Radiation on Synthetic Detergents, Sewage Colloidal Characteristics and Sewage Sedimentation, Final Report, USAEC Report COO-275, Unidynamics, Sept. 6, 1963 (*Nucl. Sci. Abstr.*, **18**: 5362).

b. Fight Water Pollution with Radiation? *Can. Chem. Process.*, **48**(11): 81-83 (1964) (summary of 6 *a*).

c. H. R. Spragg, Atomic Radiation in Sewage Processing, *Water Sewage Works*, **110**: 163-165 (1963) (same work as 6 *a*).

d. H. R. Spragg and R. L. Curtin, Ionizing Radiation in Water Reuse, *Chem. Eng. Progr., Symp. Ser.*, **63**(78): 137-142 (1967).

7. L. A. Campbell, The Use of Gamma Radiation as a Pretreatment to Chemical Precipitation in the Purification of Domestic Sewage, Canadian Report AM-6820, June 30, 1968 (this issue of *Isotop. Radiat. Technol.*).

8. The Effects of Ionizing Radiation on Colloids, USAEC Report COO-1653-15, Final Report, May 1964–November 1966, Unidynamics.

9. W. H. Burrows, W. J. Corbett, P. E. Gaffney, and C. Orr, Jr., Effects of High-Energy Ionizing Radiation on Colloidal Systems and Suspensions, Final Report, Mar. 1, 1962–Oct. 31, 1963, USAEC Report TID-20067, Georgia Institute of Technology, Engineering Experiment Station (*Nucl. Sci. Abstr.*, **18**: 12148).

10. W. M. Smith and J. O. Ledbetter, Radiation as a Possible Coagulation Aid, *Water Sewage Works*, **114**: 496-497 (1967).

11. W. N. Grune, Effects of High-Energy Ionizing Radiation on Colloidal Systems and Suspensions, *Water Sewage Works*, **112**: 262-267 (1965) (expansion of a paper presented at the Symposium on Recent Advances in Sewage Treatment, American Chemical Society, Chicago, 1964).

12. B. S. Tyler, F. J. Munno, and T. W. Cadman, Effects of Radiation on Corn Starch Sols, *Environ. Sci. Technol.*, **2**: 628-632 (1968).

13. V. I. Klassen, N. G. Malysheva, and L. P. Starchik, Radiation Effects on the Process of Sludge Precipitation (in Russian), *Tsvetn. Metal.*, **1**: 82-84 (1970) (*Nucl. Sci. Abstr.*, **24**: 19525).

14. G. M. Ridenour and E. H. Armbruster, Effect of High-level Gamma Radiation on Disinfection of Water and Sewage, *J. Amer. Water Works Ass.*, **48**: 671-676 (1956).

15. H. N. Lowe, Jr., W. J. Lacy, B. F. Surkiewicz and R. F. Jaeger, Destruction of Microorganisms in Water, Sewage, and Sewage Sludge by Ionizing Radiations, *J. Amer. Water Works Ass.*, **48**: 1363-1372 (1956).

16.*a.* C. J. Touhill, E. C. Martin, M. P. Fujihara, D. E. Olesen, J. E. Stein, and G. McDonnell, The Effects of Radiation on Chicago Metropolitan Sanitary District Municipal and Industrial Wastewaters, *J. Water Pollut. Contr. Fed.*, **41**(part 2): R44-60 (February 1969) (paper presented at the 41st Annual Conference Water Pollution Control Federation Meeting, Chicago, Sept. 22–27, 1968).

b. C. J. Touhill, E. C. Martin, M. P. Fujihara, and D. E. Olesen, Effects of Radiation on Municipal and Industrial Wastewater, 96th Annual Meeting of the American Public Health Association, Detroit, Nov. 11, 1968.

17. V. Del Vecchio, G. Branca, and S. U. D'Arca, Ionizing Radiation in Purification of Wastewater (in Italian), *Ing. Sanit.*, **1**: 20-34 (1968) [Italian Report RT/PROT-(68)15].

18. E. Gaspar and D. Serban, The Use of Nuclear Radiations in Purifying Used Water (in Romanian), *Hidroteh. (Bucharest)*, **6**: 253-256 (1961).

19.*a.* V. A. Ryabchenko, The Sanitary Significance of Colon Bacilli in Water Disinfected with Gamma Radiation (in Russian), *Gig. Sanit.*, **9**: 23-27 (1964).

b. V. A. Ryabchenko and E. L. Lovtsevich, Comparative Resistance of Enteroviruses and *E. coli* to Disinfection of Water with Gamma Radiation, *Hyg. Sanit. USSR (Engl. Transl.)*, **30**(8): 198-202 (1965).

20. V. Molinari, Resistance of *Endamoeba histolytica* in Drinking Water and Wastewater to Physical and Chemical Agents (in Italian), *Nuovi Ann. Ig. Microbiol.*, **15**: 455-462 (1965) (*Nucl. Sci. Abstr.*, **20**: 5366).

21. S. G. Sergeeva, O. V. Teshitel', and A. D. Chmyr', Bactericidal Efficiency of Gamma Irradiation of Water Used in Washing Grain (in Russian), *Izv. Vyssh. Ucheb. Zaved., Pishch. Tekhnol.*, **1**: 71-73 (1967).

22. [E. B. Fowler], Irradiating Sewage Might Improve Bacterial Nutrients, *Nucleonics*, **17**(4): 140 (1959).

23. J. O. Means, Method of Killing Organisms by Use of Radioactive Materials, U. S. Patent 3,489,218, Jan. 13, 1970 (filed Aug. 22, 1966).

24. G. I. Sidorenko, M. A. Pinigin, and V. T. Savenkova, The Use of the Combined Effect of Gamma Radiation and Chlorine for Sterilization of Waste, *Gig. Sanit.*, **28**(9): 97-98 (1963) [JPRS-21,975, pp. 10-12 (English)].

25.*a.* D. D. Woodbridge, L. A. Mann, and W. R. Garrett, Application of Gamma Radiation to Sewage Treatment, *Nucl. News*, **13**(9): 60-64 (1970) (same site described in this issue of *Isotop. Radiat. Technol.*).

b. D. D. Woodbridge, T. A. Nevin, and W. R. Garrett, Waste Treatment Process, U. S. Patent 3,539,507, Nov. 10, 1970 (filed Oct. 31, 1968).

26. M. Fleischman and R. H. Price, Gamma Irradiation of Dilute Aqueous Alkyl Benzene Sulfonate Solutions, *Environ. Sci. Technol.*, **1**: 571-577 (1967).

27. L. M. Makarochkina, A. K. Mikhailov, A. I. Gershenovich, D. I. Danilin, R. V. Dzhagatspanyan, and M. T. Filippov, Purification of Industrial Wastewaters with High-Energy Radiation (in Russian), *Khim. Prom. (Moscow)*, **45**: 905-909 (1969) (English translation: ORNL-tr-2389 by Martha Gerrard).

28. T. Sunada, Use of Radiation for Treatment of Polluted Water (in Japanese), *Genshiryoku Kogyo*, **13**(7): 34-37 (1967) (*Chem. Abstr.*, **67**: 57087w).

29. M. L. Alexander, Biodegradability of Sewage Following Gamma Irradiation, Thesis, Purdue University, 1969.
30.*a.* A. J. Condro[e]n and J. E. Etzel, Destruction of Dilute Organic Pollutants by Gamma Irradiation, *Effluent Water Treat. J.*, **7**: 528-532 (1967).
b. J. E. Etzel and A. J. Condren, Radiation Treatment of Waste Waters (abstract), *Trans. Amer. Nucl. Soc.*, **11**: 56-57 (1968).
c. A. J. Condren, Radiation-Induced Oxidation of Selected Organics in Waste Water, Thesis, 70 pp., Purdue University, 1969.
31. R. F. Byron, J. Danaczko, Jr., A. L. Dixon, and L. M. Welker (to Western Electric Co., Inc.), Radiation Decomposition of Waste Cyanide Solution, U. S. Patent 3,147,213, Sept. 1, 1964 (filed July 11, 1961).
32. G. Branca, Treatment of Waste Cyanide Solutions by Gamma Radiation, Italian Report RT/PROT-(69)23, May 13, 1969.
33. A. Yakohata and S. Tsudo, Radiolysis of Aqueous Potassium Cyanide Solution by ^{60}Co Gamma Irradiation (in Japanese), *Doitai to Hoshasen (Isotopes and Radiation)*, **4**: 393-395 (1961) (*Nucl. Sci. Abstr.*, **19**: 9184).
34.*a.* M. Steinberg, J. Pruzansky, L. R. Jefferson, and B. Manowitz, Removal of Iron from Acid Mine Drainage Waste with the Aid of High-Energy Radiation, USAEC Report BNL-11576, Brookhaven National Laboratory, 1967.
b. Ibid., Part II, USAEC Report BNL-12114, Brookhaven National Laboratory, 1968 (CONF-680536-1).
c. Ibid., Part III, USAEC Report BNL-12115, Brookhaven National Laboratory, 1968 (CONF-680536-2).
d. M. Steinberg and B. Manowitz, Purification of Mine Drainage Water by a Radiolytically Induced Chain Process, USAEC Report BNL-11220, Brookhaven National Laboratory, 1967.
e. M. Steinberg, J. Pruzansky, L. R. Jefferson, and B. Manowitz, Radiation Treatment of Acid Mine Drainage, *Trans. Amer. Nucl. Soc.*, **11**: 56 (1968).
35.*a.* E. R. Johnson, The Radiation-Induced Decomposition of Ferrous Ammonium Sulfate, *J. Amer. Chem. Soc.*, **78**: 5196-5197 (1956).
b. S. R. Huang and E. R. Johnson, *Radiation-Induced Decomposition of Some Inorganic Sulfates*, American Society for Testing and Materials, Special Technical Publication No. 400, pp. 121-138 (1966).
36. Sewage Irradiation: Is It Economical? *Nucleonics*, **16**(7): 106 (1958).
37. C. G. Bell, Sewage Treatment: Another Suggestion, *Nucleonics*, **16**(10): 116-117 (1958).
38. D. L. Narver, Jr., Is Sterilization of Sewage by Irradiation Economical? *Civil Eng.*, **27**: 54-55 (September 1967).
39. M. H. F. Friedman and C. E. Albrecht, Inactivation of Human Wastes by Ionizing Radiation, *Arch. Environ. Health*, **17**: 665-667 (1968).
40.*a.* D. M. J. Compton, S. J. Black, and W. L. Whittemore, Treating Wastewater and Sewage Sludges with Radiation: A Critical Evaluation, *Nucl. News*, **13**(9): 58-60 (1970).
b. D. M. J. Compton, W. L. Whittemore, and S. J. Black, An Evaluation of the Applicability of Ionizing Radiation to the Treatment of Municipal Waste Waters and Sewage Sludge, *Trans. Amer. Nucl. Soc.*, **13**: 71-72 (1970).
c. W. L. Whittemore, D. M. J. Compton, and S. J. Black, Ionizing Radiation for the Treatment of Municipal Waste Waters, Final Report, Feb. 26, 1969–Feb. 28, 1970, USAEC Report GA-9924, Gulf General Atomic Incorporated, February 1970.
41.*a.* Report prepared for the Metropolitan Sanitary District of Greater Chicago by the Bechtel Corporation, April 1968.
b. Sewage Irradiation, *Washington Sci. Trends*, **21**(10): (Dec. 16, 1968).
42. W. M. Jackson, Radiation Processing of Poultry Wastes, *Isotop. Radiat. Technol.* **8**(2): 223-225 (Winter 1970–1971) (USAEC file No. NP-18048, 1969).
43. K. J. Mitchell (to General Electric Company Ltd.), Improvements in or Relating to Methods and Apparatus for Treating Sewage, British Patent 1,079,632, Aug. 16, 1967 (filed Mar. 30, 1965).
44. N. E. Nielson, Aqueonics Inc., personal communication, Jan. 18, 1971.
45. A. I. Mytelka, Radiation Treatment of Industrial Wastewaters: Economic Analysis, *Chem. Eng. Progr., Symp. Ser.*, **65**(97): 246-250 (1969) (Aerochem Report TP-188a, June 1969).

Note added in proof: After this article was prepared for the printer, a bibliography, Radiation Treatment of Sewage, compiled by E. A. Newland, September 1970 (Australian Report AAEC/LIB/Bib-254), was received. Significant reports appear both in the Australian bibliography and in the reference list for this article, but the latter is more up to date.

Conceptual Design of an Irradiation Test Facility for Waste Water and Sewage Sludge*

Summarized by Martha Gerrard

*Report prepared for the Metropolitan Sanitary District (MSD) of Greater Chicago by the Bechtel Corporation, April 1968. The editors of *Isotopes and Radiation Technology* thank Dr. David T. Lordi, Acting Director of Research and Development of MSD, for making the report available and for reviewing this summary.

Supplementary Keywords: *water and waste treatment; facility description; source, gamma; machine irradiation; comparison; irradiator design; pilot-plant scale; ^{60}Co.*

Abstract. *The conceptual design of a pilot plant for radiation processing of sewage is described briefly.*

XUM

The report summarized here presents a conceptual design and order-of-magnitude cost estimates for a test facility to irradiate waste waters and sewage sludges. Although, for design purposes, ^{60}Co was selected as the radiation source, cost estimates were also prepared for electron accelerators as the source of radiation. The design and cost estimates were made by the Bechtel Corporation at the request of the Metropolitan Sanitary District of Greater Chicago (MSD), whose research had indicated that a radiation dose of 250 krads disinfects sludge, promotes settling of waste-activated sludge, and accelerates digestion.

The selection of ^{60}Co as the radiation source was based on its ready availability in the quantities required for large-scale use. Encapsulation techniques for this radionuclide are already well developed, and the feasibility of using ^{60}Co gamma radiation as a sterilizing agent has been demonstrated in the half dozen or so commercial plants in the United States and Europe which use it for sterilization of medical supplies. The electron beam from a linear accelerator was a secondary choice because it would probably produce results similar to those of gamma radiation, although this has not yet been demonstrated. If the accelerator should prove satisfactory, it might be cheaper than ^{60}Co. Some other radiation sources that might be suitable are ^{137}Cs, spent power-reactor fuel elements, and a nuclear reactor. However, ^{137}Cs is not yet available on a large scale nor are encapsulation facilities adequate. Spent power-reactor fuel elements present handling problems, and the possibility of fission-product leakage must be considered. A nuclear reactor was disqualified chiefly because a reactor could not be built and licensed in less than 3 or 4 years. However, it should be given serious consideration in any future designing of large-scale sewage-processing facilities.

DESIGN

The conceptual design of the sewage-processing facility includes an irradiator (Fig. 1), a pool shield, concrete shielding, pumps, piping, and the usual utilities, offices and changerooms, heating and ventilation, lighting, radiation-monitoring equipment, emergency dumping facilities, etc. The nominal capacity of the test plant is 2×10^6 gal/day, to which the 3.92-MCi source supplies a dose of 50 (±10%) krads. The irradiator efficiency is 76.5%. The ^{60}Co must be replaced at a rate of ~12% per year to maintain the energy output at the initial level.

Streams to be processed flow by gravity to the test facility. Because of the experimental nature of this facility, provision was made for both raw sewage and various intermediate sludge streams to be diverted to the irradiator. The irradiated effluent from the test facility is routed to a new concentration tank for observation and sampling, and from here it can be sent back to the digesters, to other concentration tanks, to the main drain, or again to the irradiation facility for additional exposure. Provision was also made to permit variation, within limits, of the radiation dose to the sewage stream. Higher doses can be administered by longer residence of the sewage stream in the irradiator, achieved either by a slower flow rate or by batch recycle.

The irradiator is located in a dry well that opens off the bottom of the shielding pool, a 1.5-in.-thick stainless-steel plate, from which the irradiator tank hangs. A tube sheet covering the irradiator is bolted to this steel plate, and a gasket prevents contact of the process stream with the water in the pool. In the tube sheet—a hexagon, 5 ft 8 in. across the corners—are welded tubes to contain the ^{60}Co. These project into the fluid being processed when the tube sheet is in place. Each tube has a 0.555-in. inside diameter and a 0.035-in.-thick wall, is 13 ft 2 in. long, and has a plugged bottom end. The top of each terminates with a 3-in.-diameter tapered flare or cone, through which the cobalt is inserted or removed.

The fluid being processed flows through an inner region formed by a baffle in the tank. The baffle, made of 0.140-in.-thick stainless-steel sheets welded to the bottom plates, guides the fluid as it flows downward during its period of maximum radiation dose.

The pool itself is 15 ft deep by 20 ft wide by 30 ft long. It is completely lined with type 304 stainless steel, and the walls are further lined with 0.140-in.-thick stainless-steel sheets and the floor with ¼-in.-thick stainless steel. The water in the pool is filtered so that handling operations can be easily observed. A deep well is provided for storage of the source when the pool must be drained for any reason.

RADIATION SOURCE

The ^{60}Co is in the form of cylinders, called "pencils," which are available commercially (Fig. 2). Six pencils are stacked in each tube, a total of 1302 pencils in the 217 tubes. Each pencil contains a solid metal rod or small particles of cobalt metal wire. The pencils are about 0.250 in. in inside diameter and

XUM

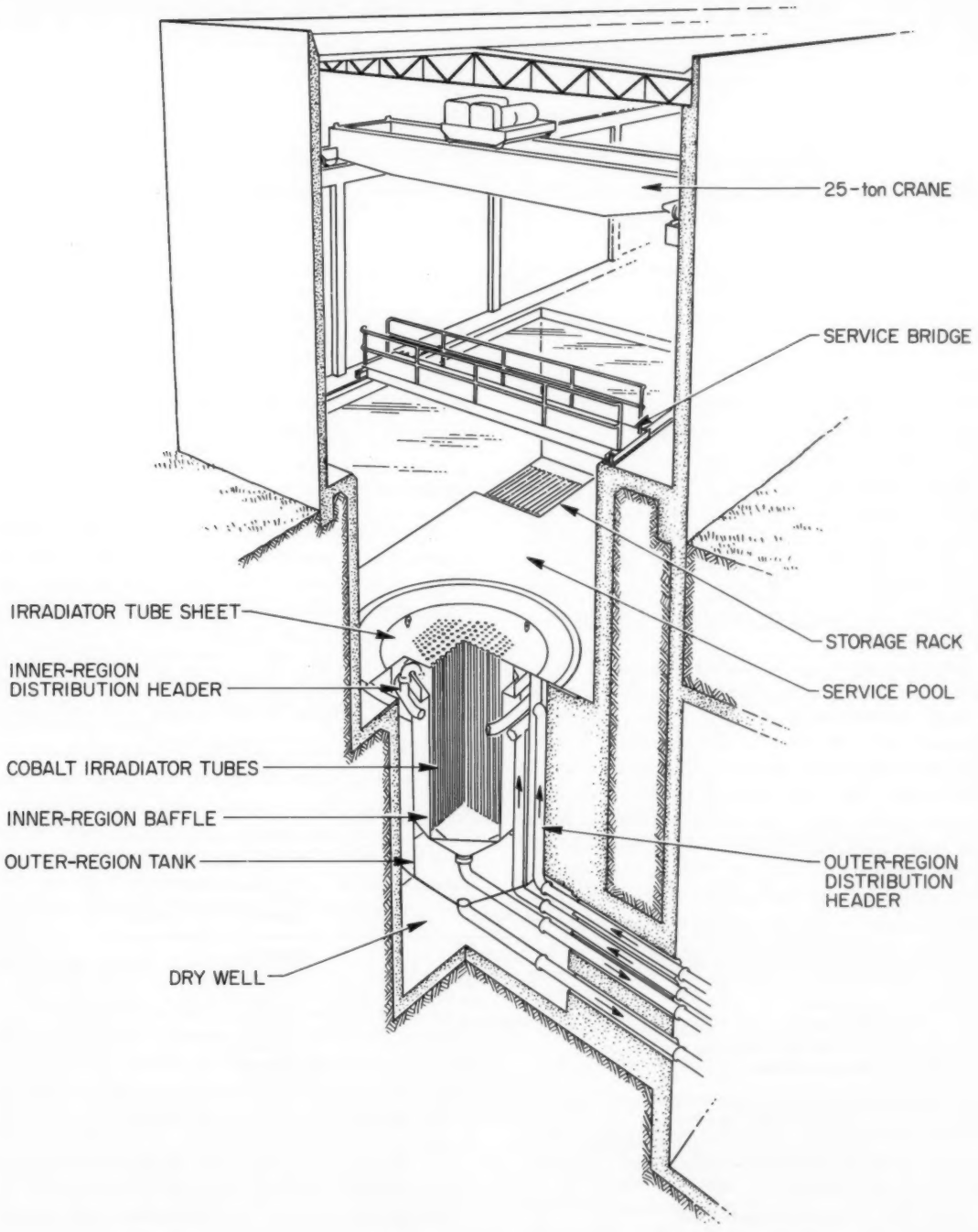

Fig. 1 Cutaway perspective through ^{60}Co irradiator and service-pool building (conceptual design).

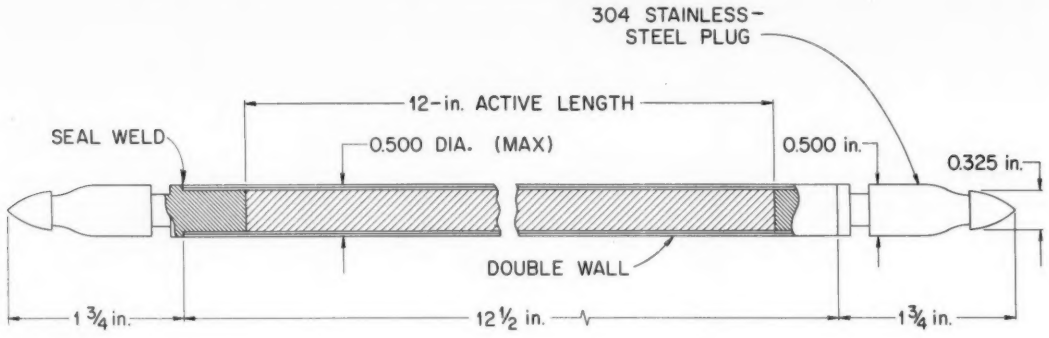

Fig. 2 Typical ^{60}Co pencil.

16 in. long, with an active length of 12 in. End fittings are provided for attachment of tools when the pencils are to be handled. The pencils are doubly encapsulated in 0.020-in.-thick stainless steel, so that there are three barriers between the fluid being processed and the radioactive material—the two wall thicknesses of the double encapsulation and one wall thickness of the thimble. Decay heat—1 W per 60 Ci of activity (50 W per pencil)—is removed by water in the tubes, around the pencils.

The dose rate is 3.65 Mrads/hr in the section within the hexagonal flow baffle where the source is located. A slight nonuniformity of dose is not considered important since the material being irradiated is fluid and its mixing by air agitation should ensure an essentially homogeneous dose.

COST

The ^{60}Co test facility itself could be constructed in 9 months at an estimated cost of $\$2.7 \times 10^6$ (Table 1). With the initial cost of the ^{60}Co, based on quotations from suppliers, taken as $\$2.6 \times 10^6$, with no allowance for salvage of partially decayed ^{60}Co, the total facility would cost around $\$5.3 \times 10^6$. The complete linear accelerator facility was estimated to cost about 20% less. These estimates do not include cost of licensing the facility, land, or conceptual design.

Table 1 Sewage-Irradiation Facility

	^{60}Co		Linear electron accelerators*	
	Amount	Total	Amount	Total
Civil and structural engineering	$1,244,000		$805,000	
Piping	263,000		255,000	
Electrical	189,000		244,000	
Mechanical	189,000		78,000	
Heating and ventilating	46,000		37,000	
Instrumentation	27,000		22,000	
Total specific construction cost at March 1968 price list		$1,958,000		$1,441,000
Engineering, procurement, estimating, and related home office services		300,000		300,000
Contingency		451,000		348,000
Total constructed cost		$2,709,000		$2,089,000
Radiation source		2,600,000		1,980,000
Total project cost		$5,309,000		$4,069,000

*Three units, 30-kW electron beam, 5-MeV power.

Biological–Gamma-Radiation System for Sewage Processing

By Leland A. Mann*

Supplementary Keywords: *water and waste treatment; pollution; industry, utility; source, gamma; economics; microbiology; sterilization; cobalt-60.*

Abstract: *A biological–gamma-radiation method for purifying sewage at a cost competitive with that of conventional advanced treatments was developed, and a pilot plant and a commercial plant were designed, constructed, and tested. Settleable-solids concentration and turbidity are decreased, and a satisfactory biological oxygen demand (BOD) is maintained. Important improvements that are not economically feasible with conventional treatments alone are the destruction of almost all coliform bacteria, of some viruses, and of more than 90% of both biodegradable and nonbiodegradable detergents, with indication that algae do not grow in the effluent. Plants already in existence could be modified by the addition of irradiator and filter modules to increase system capacity and improve effluent quality.*

This article describes the development of a practical, tested, and cost-proven method of sewage treatment in which gamma irradiation is a key step. Patents have been applied for in the United States and six other countries. Two U. S. patents have been awarded, and one has been issued.[1]

A 50-krad dose tested in the pilot plant, and shown to be effective, was used as the design dose for the commercial plant. The indications in the literature that higher doses are necessary suggest that, in the earlier work, the importance of two crucial factors may have been overlooked, that is, uniformity of dose to the fluid and efficiency of utilization of the radiation.

PILOT PLANT

The pilot-plant design was a compromise among cost, uniformity of dosage, degree of utilization (90% or more) of the gamma energy, and plant capacity (360 gal/day) adequate for providing information for scale-up to a commercial system. The size of the ^{60}Co source used was based on the results of preliminary tests, which had shown 50 krads to be adequate.

The pilot-plant irradiator unit, which was designed, constructed, and licensed to serve also as the ^{60}Co transportation cask, was loaded at Lockheed-Georgia Nuclear Laboratory, Dawsonville, Ga., with 560 Ci of doubly encapsulated ^{60}Co strips of Brookhaven National Laboratory design. The unit was installed at the University Center for Pollution Research at the Florida Institute of Technology, Melbourne, Fla., on Mar. 26, 1968.

After addition of appropriate tanks, instrumentation, plumbing, and auxiliary equipment, the plant was tested at several flow rates, including the nominal flow rate of 0.25 gal/min, on normal sewage obtained from the city of Melbourne, Fla. In these tests almost all coliform bacteria were killed, and both biodegradable and nonbiodegradable detergents were more than 90% destroyed. Limited tests at the design flow rate, i.e., to give a 50-krad dose, on Parathion in water resulted in 25 to 35% destruction of this insecticide. Other pesticides and herbicides have not been studied in the equipment.

PALMDALE PLANT

As a result of the favorable findings obtained in the pilot plant, Energy Systems, Inc., built, and is operating, for Lykes Brothers Inc., Tampa, Fla., a commercial treatment plant located at Fisheating Creek Campground near Palmdale, Fla., west of Lake Okeechobee. This nominal 10^4 gal/day plant has operated continuously since startup on Mar. 10, 1970. The actual flows experienced have been from less than 10^3 gal/day to more than 1.5×10^4 gal/day.

Construction Features

The plant consists of several modules (Fig. 1):

Wet well. The conventional collection tank has a rough screen, two centrifugal pumps, level controls, and aeration and partial-recirculation systems.

Biological treatment unit. The biological treatment unit is a conventional 10^4 gal/day extended aeration plant, with volume-flow control at the inlet and a screen filter in the effluent pipe.

Irradiator. Externally, the irradiator is approximately 14 ft square by 9 ft high. It is constructed of

*Energy Systems, Inc., 150 Wickham Road, Melbourne, Fla. 32901.

XUM

high-grade reinforced concrete, through which influent and effluent pipes follow spiral paths to the core. The minimum concrete thickness in any direction from the core is 50 in. The top shield is made of three square slabs of stepped sizes, each fitted with lifting rings.

Palmdale plant empties into a cypress-swamp slough, which is dry most of the year, and finally into Fisheating Creek at a point several hundred yards from the effluent outlet, and downstream of the campground.

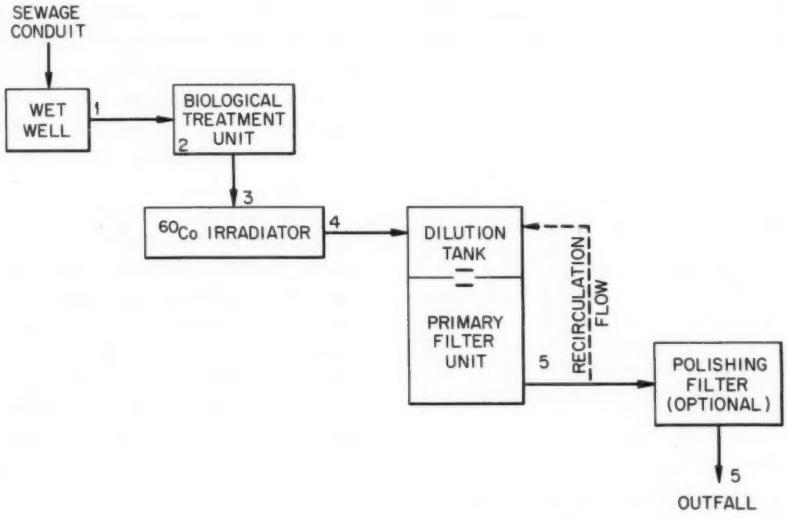

Fig. 1 Schematic flow diagram of Palmdale sewage-processing plant. Numbers indicate sampling points.

Sixteen thousand curies (as of May 1, 1969) of ^{60}Co was charged into the irradiator in the design pattern on Aug. 1, 1969. The core design, which is proprietary, produces maximum feasible uniformity of irradiation of the fluid and allows less than 10% waste of gamma-radiation energy by absorption in the concrete shield. Flow from the extended aeration plant, through the irradiator, to the filter dilution tank is entirely by gravity—there are no moving parts in the irradiator module. A gamma dosimeter of 200 mR capacity is installed in contact with the effluent pipe from the irradiator, inside a Cyclone security fence.

Primary filter system. The effluent flows from the irradiator to a 10^3 gal dilution tank and then through a parallel pair of vacuum-operated diatomaceous-earth leaf-type filters. It is then pumped through a proportioning valve, which returns a controllable fraction of the flow to the dilution tank.

Activated-charcoal (polishing) filter. The activated-charcoal filter, which is optional, is used to treat the effluent from the primary filter system before discharge to the environment. The effluent of the Multiplication of bacteria and other contaminants entering the filter systems (from the air, filter mediums, etc.) is prevented by chlorination of the circulating liquid to 0.5 ppM residual chlorine; this level of chlorine has been found adequate. The quantity of chlorine required is considerably less than that used by typical conventional plants.

Improvements in Water Quality

Irradiation of sewage to a nominal 50-krad dose in the Palmdale plant has revealed several significant chemical, physical, and biological accomplishments.

Chemical and Physical Improvements. Figure 2 shows representative results of analyses (performed by the methods of Ref. 2) of samples taken at the locations indicated in Fig. 1. As sewage passed through the irradiator (sampling positions 3 and 4), the dissolved oxygen (DO) content decreased from 5.1 to 3.2 ppM. These values are misleading, however, since the irradiation converts some oxygen to peroxide and ozone and the oxidation potential actually increases, as

XUM

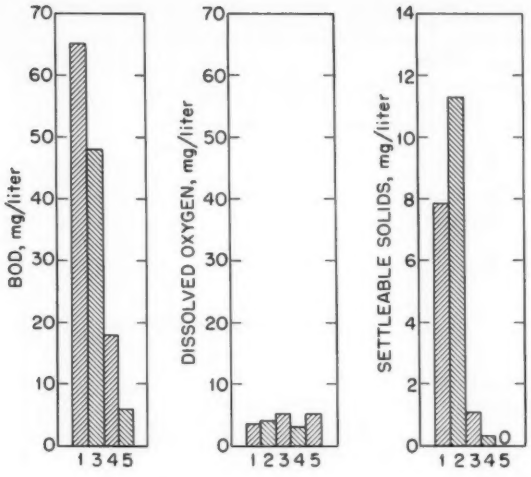

Fig. 2 Average analyses at various locations in the Palmdale plant: (1) effluent from wet well; (2) dip sample, basic treatment unit; (3) influent to irradiator; (4) effluent from irradiator; and (5) final effluent.

shown by the BOD decrease from 48 to 17.5 ppM. The settleable-solids concentration was decreased approximately 75%; the turbidity was decreased 23% (from 26.2 to 20.2 units); and the *p*H increased from 6.1 to 6.5. Biodegradable and sodium lauryl sulfate detergent contents were decreased more than 90%.

With the addition of a primary filter-system module to the irradiator effluent, settleable solids (sampling location 5) were decreased to zero and the DO increased to 5.3 ppM. Both the BOD and the turbidity were further decreased to 6 ppM and 15 units, respectively.

The addition of an activated-carbon (polishing) filter module to the irradiator–primary filter system completely decolorized the effluent and decreased the turbidity to nearly zero.

Biological Improvements. The 50-krad dose had a marked effect on the microbiological composition of the sewage. All *Escherichia coli* bacteria were killed, and coliform bacteria in general were decreased in number by a factor of 10^4 or more. No facilities were available for testing virus kills, but the literature[3] leaves no reasonable doubt that poliomyelitis, influenza, and many other viruses are either destroyed or drastically decreased in number with the dosage used. Destruction of viruses by conventional methods of water treatment is not economically feasible.

Algae were added to a few samples taken simultaneously from locations 3 (conventional plant effluent) and 5 (after irradiation and primary filtration). In each case, an algae bloom resulted in the conventional plant effluent, but no bloom or growth of algae occurred in the irradiated fluid. Further studies of this phenomenon are planned because of the obvious importance for decreasing algae in streams.

Dependence on Flow Rate. A very interesting fact became evident in the early tests of parameters in the operation of the Palmdale plant. When the flow rate was increased from the nominal 10^4 to 2×10^4 gal/day, there was no measurable decrease in bacteria kill or change in the other measured parameters (BOD, settleable solids, DO, *p*H, etc.), although the quality of the effluent entering the irradiator from the extended aeration module had deteriorated badly. An obvious implication is that capacities of existing secondary sewage-treatment plants can be greatly increased by simply adding a Palmdale-type irradiator and filter system. A parallel implication is that at least a partial treatment could be provided when there are operating difficulties in the conventional plant, thus eliminating the all-too-prevalent practice of bypassing sewage around the treatment plant in such cases.

COSTS*

The cost of water treatment must not be considered without attention to water quality. Since the Palmdale treatment plant is based on the modular concept, water quality as well as plant cost depends on the modules selected.

The quality requirements of the purchaser are determined, and the modules required to obtain the specified quality are selected. The purchaser chooses the final effluent quality after considering the costs and advantages of each module. For example, a plant whose effluent is to be used for irrigation might omit all treatment after irradiation and thus furnish some nutrient for the plant life to be irrigated. Water for some industrial processes could omit the polishing-filter module if water color is not objectionable. Recreational water, water to be recycled to the inlet of a normal municipal freshwater treatment plant, and potable water for direct distribution would each have

*Since this paper was prepared, further studies have shown that costs may be reduced by 10 to 15% by using smaller quantities of ^{60}Co but with more frequent replenishments to maintain the same minimum dosage.

XUM

different requirements. The analytical results shown in Fig. 2 can be used as a preliminary guide to modules needed for the desired quality.

Estimated capital costs for addition of irradiator plus primary filtration modules to conventional treatment plants and operating costs are shown in Figs. 3 and 4 and Table 1. Interest cost is not included in the estimates because the rate is essentially the same for the new system as for conventional systems.

The difference between capital construction costs and total costs as functions of plant capacity is that of operating and maintaining the system in the usual sense plus the cost of additions of ^{60}Co at 4-year intervals to restore the radiation intensity to its initial level. This ^{60}Co replenishment schedule is an economic compromise between the handling cost of frequent additions and the amount of activity added and is subject to revision as the costs of cobalt and cobalt handling change.

If the cost of conventional treatment, without chlorination costs, is added to the costs shown in Table 1 and Figs. 3 and 4, the cost of a treatment plant consisting of a conventional secondary treatment plus irradiator and primary filtration modules is found to be less than that of conventional advanced-treatment plants.[4]

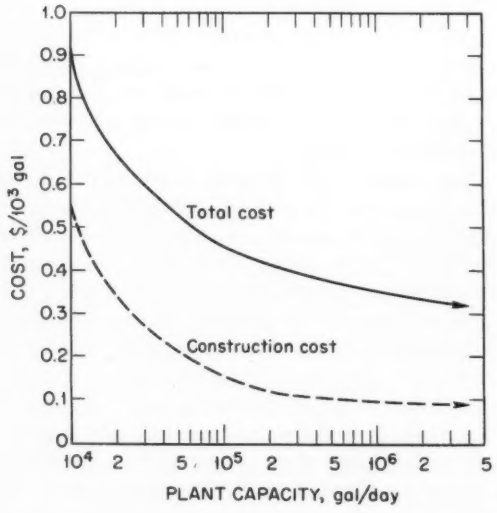

Fig. 3 Construction and total costs vs. plant capacity for the addition of irradiator and filter system to conventional sewage-treatment plants. (Based on assumption of 40-year amortization and $0.35/Ci ^{60}Co.) Difference between two curves is maintenance, operation, and periodic ^{60}Co-makeup costs.

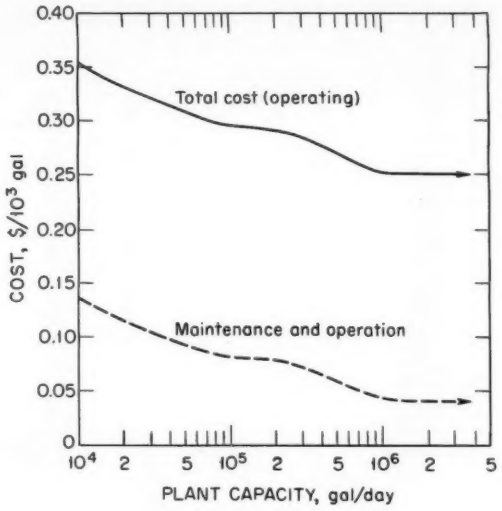

Fig. 4 Maintenance and operation costs and total operating cost for additions of irradiator and filter system to conventional sewage-treatment plants. (Based on $0.35/Ci ^{60}Co.) Difference between curves is periodic ^{60}Co-makeup costs.

Cobalt-60 Costs

The curies of ^{60}Co required in the initial charge to deliver the nominal (5×10^4 rads) dose can be calculated as follows:

$$\text{Ci} = \frac{1.48FKe^{\lambda\eta}}{E}$$

where 1.48 is a constant; F = plant capacity in gal/day; K = minimum acceptable dose in rads, divided by 5×10^4 rads; E = fraction of gamma energy absorbed in the fluid; λ = decay constant for ^{60}Co (0.1315 per year); and η = interval, in years, between ^{60}Co additions.

A reasonable approximation in which $K = E$ and $\eta = 4$ years is

$$\text{Ci} = 1.48e^{\lambda\eta}F = 2.50F$$

The cost of ^{60}Co for the initial charge would then be $2.50FP$, where P is the cost per curie of ^{60}Co. The cost of ^{60}Co for each makeup addition is

$$1.48e^{\lambda\eta}FP(1-e^{-\lambda\eta})$$

or $1.02FP$ for $\eta = 4$.

XUM

Table 1 Capital Cost Vs. Plant Capacities*

Plant capacity, 10^3 gal/day	Cost (in thousands)			Unit cost per 10^3 gal
	Construction	^{60}Co (at \$0.35/Ci)†	Total	
10	\$ 65	\$ 17	\$ 82	\$8200
50	101	56	157	3140
100	128	106	234	2340
250	176	255	431	1720
500	296	510	806	1610
750	402	762	1164	1550
1000	536	1015	1551	1550
3000‡	1301	2770	4071	1360

*To convert any of the above costs to cents/10^3 gal over a 40-year amortization period (excluding interest), multiply total cost in dollars by 6.85, and divide results by the capacity in gal/day.

†Includes transportation and loading costs.

‡The cost of the 3×10^6 gal/day plant was based on the use of an ocean outfall system.

Total Irradiator Costs

The total cost per 10^3 gal for the irradiator part of an overall system can be expressed over the entire lifetime of the plant as

$$\frac{2.74}{NF}(C_1 + C_2 + C_3 + C_4)$$

where 2.74 is a factor that puts costs per 10^3 gal on a yearly basis (10^3/365 days per year); N = lifetime of plant in years; C_1 = total cost of ^{60}Co (FOB supplier); C_2 = total cost of ^{60}Co transportation and additions; C_3 = total cost of initial construction, other than ^{60}Co costs; C_4 = costs of operation and maintenance, other than ^{60}Co costs.

The ^{60}Co purchase cost (FOB supplier) per 10^3 gal is, then,

$$\frac{2.74}{NF}(C_1) = \frac{4.06KPe^{\lambda\eta}}{EN}\left[1 + \left(\frac{N}{\eta} - 1\right)(1 - e^{-\lambda\eta})\right]$$

Where $N = 40$, $\eta = 4$, and $K = E$, this equation becomes

$$\text{Cost}/10^3 \text{ gal} = 0.800P$$

Item C_3 is the sum of costs of the core of the irradiator, the high-quality reinforced-concrete shield, the Cyclone security fence, and the instrumentation for monitoring effluent activity. Item C_4 is the very small operation and maintenance cost for the irradiator module in addition to that for the conventional modules. This cost consists almost entirely in inspecting the radiation monitor and recording the readings, and only an occasional routine cleaning and painting since there are no moving parts involved. No additional personnel are required.

The unit cost of ^{60}Co is a major consideration, although not decisive, in the competitive cost comparison with conventional (nonirradiative) advanced water-treatment systems designed to produce comparable quality effluent.

All costs of conventional and nonconventional systems are, of course, estimates with varying confidence levels. The cost estimates used for bionuclear-system plants are based on actual costs of the Palmdale plant, with adjustments for differences in capacities, and therefore are relative to costs in Florida. It seems reasonable to assume that comparative costs between bionuclear and conventional systems would not vary widely with location, at least in the United States.

RADIATION SAFETY

Cobalt-60 irradiation causes no residual radioactivity in the effluent. Ordinary high-quality reinforced-concrete shielding of adequate thickness and properly prepared, poured, set, and moistureproofed by accepted methods is a completely satisfactory shield against ^{60}Co radiation. Each additional 8-in. thickness of concrete provides shielding for a 10-fold increase in

XUM

source strength and gives, conservatively, adequate protection at minimum cost.

The ratio of ^{60}Co signal to natural-background environmental radiation at the surface of the Palmdale irradiation shield is too low to be measured—only the natural background can be detected. The Cyclone security fence, 10 ft high and topped with barbed wire, will be maintained around the irradiator as an additional safeguard. The fence also prevents vandalism to the monitoring instrumentation. No evidence of corrosion of the double-stainless-steel encapsulations of ^{60}Co has appeared. Although no corrosion was expected, monthly radiation tests of effluent to ensure continued integrity of the encapsulation will be made. (REG)

REFERENCES

1. D. D. Woodbridge, T. A. Nevin, and W. R. Garrett, Assignors to Energy Systems, Inc., Waste Treatment Process, U. S. Patent No. 3,539,507, Nov. 10, 1970 (filed Oct. 31, 1968).
2. *Standard Methods for the Examination of Water and Wastewater Including Bottom Sediments and Sludges,* prepared and published jointly by American Public Health Association, American Water Works Association, and Water Pollution Control Federation, 12th ed., American Public Health Association, Inc., New York, 1965.
3. J. Kivel, M. S. Chatterton, and R. J. F. Palchak, Microbial Inhibition Systems Employing Radiation (MISER), USAEC Report NYO-3736-2, Atlantic Research Corporation, November, 1966.
4. D. G. Stephan and L. W. Weinberger, Wastewater Reuse—Has It "Arrived"?, *J. Water Pollut. Contr. Fed.*, **40**: 529-539 (April 1968).

Radiation Treatment of Industrial Waste Waters: An Economic Analysis*

By A. I. Mytelka†

Supplementary Keywords: *water and waste treatment; source, gamma; radiolysis; comparison; economics; cobalt-60.*

Abstract: *An economic analysis of radiation treatment of industrial waste waters shows that overall oxidation-reaction efficiencies must be increased by factors varying from 2 to 25 times those presently obtainable, depending upon the waste considered and the allowable treatment cost, if radiation treatment is to become economically feasible.*

Elimination of water pollution is dependent upon the development and application of new economical waste-treatment processes and techniques. In response to this need, researchers are currently evaluating various advanced chemical and physical treatment methods, such as (1) aqueous oxidation by inorganic oxidizers, (2) electrodialysis, (3) reverse osmosis, (4) freezing, and (5) radiation treatment.

Radiation treatment of industrial wastes has been a part of Aerochem's advanced waste-water investigation for several years. The objectives of the program are to make a technical and economic evaluation of the use of radiation to oxidize organics in aqueous solution and to increase the biodegradability of refractory industrial wastes prior to their discharge into industrial or municipal biological waste-treatment plants. A flow scheme envisioned for pretreatment of industrial effluents is shown in Fig. 1. Biodegradable effluents from processes B, C, and D can flow directly into the waste-treatment plant. However, effluent from process A is not biodegradable; it would be diverted (as shown by the dotted lines) to a pretreatment facility where the organics in the water would be rendered biodegradable. Afterward, the pretreated effluent would enter the biological treatment plant.

Besides ionizing radiation, numerous means of oxidation can be used in this pretreatment step—direct electrochemical oxidation, ozonization, and oxidation with permanganate or catalyzed hydrogen peroxide. The economics of many of these processes for treating sewage effluents have been summarized elsewhere[1,2] with the conclusion that they are not yet economically feasible. In this article, the economics of using ionizing radiation as a pretreatment method for increasing the biodegradability of industrial wastes are examined.

*An edited version of Aerochem Report TP-188a, June 1969.

†Assistant Chief Engineer, Interstate Sanitation Commission, New York, N. Y.

XUM

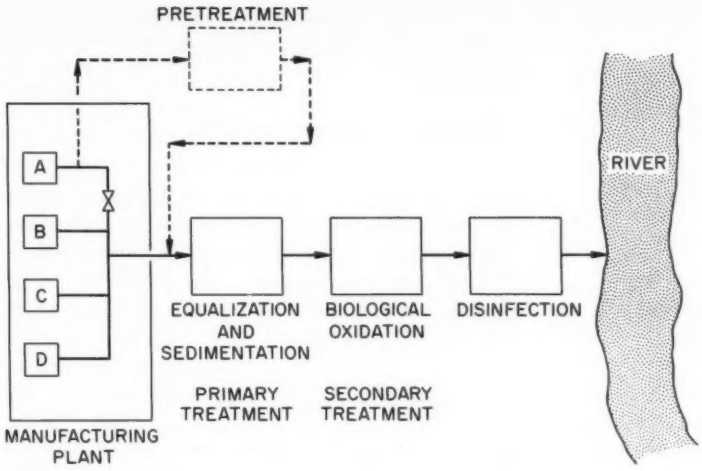

Fig. 1 Typical biological waste-treatment flow system.

ECONOMIC ANALYSIS

The method used to analyze the economic feasibility of radiation treatment of industrial wastes was to (1) assume a range of costs of treatment ($/10^3 gal), (2) design and estimate the cost of a plant to provide treatment at these costs, (3) calculate a reaction rate that would allow the use of radiation treatment to meet each cost, and (4) compare the calculated reaction rate to that obtained in the laboratory.

Allowable Cost of Treatment

The cost of treatment that industry will be able to afford is difficult to predict. Obviously, each company must be able to afford whatever its competitors can in order to stay in business. The allowable cost of waste treatment is related to the cost of process water: the higher the cost of process water, the larger the allowable cost of waste treatment prior to recycling the water for process use. With these facts in mind, waste-treatment costs between $0.20/10^3 gal and $1.00/10^3 gal seem to be reasonable; this range of values was used in the calculations.

Plant Design and Costs

The plant design and costs are divided into two parts: (1) energy source, i.e., ^{60}Co, and (2) the rest of the plant. The cost of part 1 is a function of the quantity of ^{60}Co needed and its unit cost. The cost of part 2 is fixed by the plant flow, which is assumed to be 1 million gal (10^6 gal) per day with a detention time of 1 hr.

Energy Source. The ^{60}Co cost is determined as a function of the initial investment cost [(Ci of ^{60}Co needed)($/Ci)]: interest at 6%, plus cost of replacement cobalt (12.5% of initial investment), plus freight and handling costs (2.5% of initial investment). The annual ^{60}Co cost is summarized by:

$$\text{Cost, \$/10}^3\text{ gal} = \frac{(\text{Ci needed})(\$/\text{Ci})(0.21)}{3.65 \times 10^8\text{ gal}} \qquad (1)$$

Plant. The cost of the rest of the plant will depend upon many factors, including the actual plant design. For present purposes, the flow-through radiation pretreatment plant is assumed to consist of two concentric cylinders (Fig. 2). The ^{60}Co will be contained in the inner cylinder, and the volume between the cylinders will contain the effluent to be treated. In this manner, the effluent will act as a liquid radiation shield and minimize other shielding necessary for the protection of operating personnel. The total cost of the plant and associated accessories is computed, as shown in Table 1, to be $150,000, but a total cost of $200,000 is assumed so that the estimate will be conservative. The annual cost, which includes depreciation over a 20-year period plus operating costs, will be:

Interest (6%)	$12,000
Depreciation (5%)	10,000
Maintenance*	5,000
Power*	2,000
*p*H adjustment*	7,300
	$36,300

*Estimate.

XUM

Dividing the annual cost by the yearly volume treated gives the cost per unit volume:

$$\text{Cost, \$/10}^3\text{ gal} = \$36{,}300/3.65 \times 10^8\text{ gal} = \$0.10/10^3\text{ gal} \quad (2)$$

The two costs, from Eqs. 1 and 2, can be combined to give the overall annual treatment costs:

$$\text{Cost, \$/10}^3\text{ gal} = \$0.10/10^3\text{ gal} + \frac{(\text{Ci needed})(\$/\text{Ci})(0.21)}{3.65 \times 10^8\text{ gal}} \quad (3)$$

Values of Eq. 3 have been calculated and are shown graphically in Fig. 3. Three different prices for ^{60}Co or its equivalent in the form of spent fuel elements were chosen. The \$0.35/Ci price is an approximate current price; \$0.25/Ci is a probable near-future price; and \$0.10/Ci is an optimistic long-range price. Figure 3 shows that, at the present time, a \$0.20/10^3 gal maximum treatment cost would impose a ceiling of 0.5 MCi of ^{60}Co that could be afforded for a radiation facility, while a \$1.00/10^3 gal maximum treatment cost would allow the use of 4.5 MCi of ^{60}Co. If ^{60}Co decreases in price to \$0.10/Ci in the future, 1.7- and 15.5-MCi facilities would be economically feasible at \$0.20/10^3 gal and \$1.00/10^3 gal treatment costs, respectively.

Calculated Overall Reaction Rates

For a 10^6 gal/day flow rate and 1-hr detention time, the liquid volume being irradiated is fixed at 5500 ft^3. Based upon the radiation dose obtainable from the quantity of ^{60}Co that can be afforded for various radiation-treatment costs (Table 2), overall oxidation efficiencies for different amounts of oxidation were calculated from:

$$G_{-O_2 eq} = 0.128 \times \Delta COD/M \quad (4)$$

where $G_{-O_2 eq}$ is oxidation efficiency,* COD is chemical oxygen demand in mgO_2/liter, and M is radiation dose in Mrads. Results are presented graphically in Fig. 4. This figure indicates that if cost is, for

*G is defined as the number of molecules formed or destroyed per 100 eV of energy absorbed. Since the COD is measured in terms of oxygen equivalents, $G_{-O_2 eq}$ is defined as the change in COD in mgO_2/liter per 100 eV of energy absorbed.

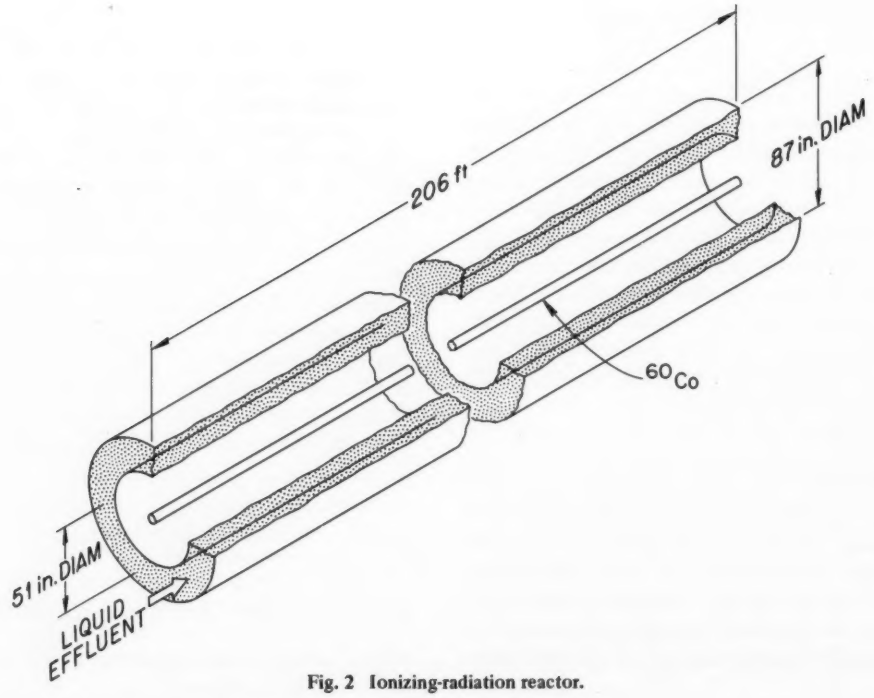

Fig. 2 Ionizing-radiation reactor.

Table 1 Plant Cost Breakdown

Two concentric steel reactor cylinders*		$ 37,800
Support for inner cylinder		1,200
Inner tube to hold ^{60}Co pencils		2,000
		$ 41,000
Cobalt-transfer machinery		35,000
Radiation-monitoring system		4,000
*p*H-adjusting equipment and controls		5,000
Aeration equipment		5,000
Miscellaneous		10,000
	Subtotal	$100,000
Installation and design		50,000
	Total	$150,000

*One 51 in. in diameter, the other 87 in. in diameter, both 0.25 in. thick and 206 ft long.

instance, limited to $0.50/10^3 gal for radiation treatment, and if the amount of pretreatment needed is, for example, an oxidation equivalent to a decrease in the COD of 100 mgO_2/liter, then, at the present time, this process would be economically feasible if the oxidation reaction has an overall G_{-O_2eq} value of 190 or more. If the ^{60}Co cost drops to $0.10/Ci, then—with the same allowable treatment cost—a G_{-O_2eq} value of only 52 will be satisfactory.

Comparison of Overall Reaction Rates

The data in Table 3 show increased biodegradability of two wastes resulting from radiation pretreatment. The COD/BOD ratio of about 4 after radiation pretreatment indicates that the biodegradability of these samples is approaching that of municipal sewage. Assuming that this biodegradability of the wastes is satisfactory, the overall G_{-O_2eq} that must be reached to make radiation treatment economically feasible for the above wastes is obtained from Fig. 4 by using the COD decrease value from Table 3. The value of the ordinate (G_{-O_2eq}) on Fig. 4 where the COD value intersects the allowable treatment cost is the minimum overall oxidation efficiency that can be tolerated, and this value is considerably larger than the experimental

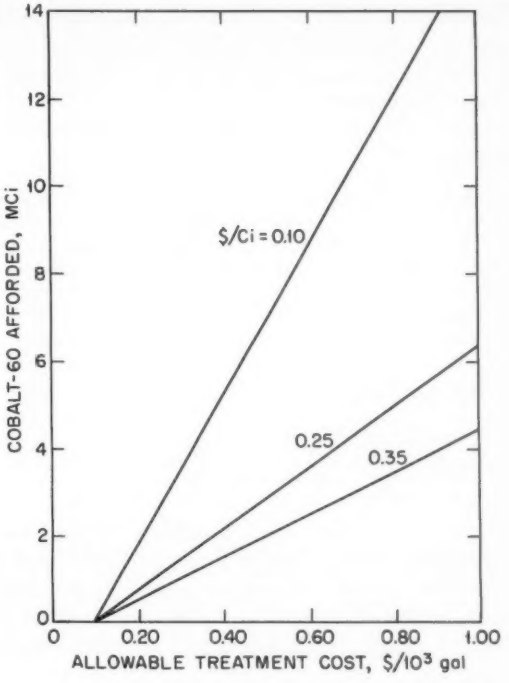

Fig. 3 Quantity of ^{60}Co that can be afforded for different treatment costs for a 1-Mgd treatment plant.

Table 2 Calculation of Dose to Waste from Exposure to ^{60}Co for 1 Hr

(1) Assumptions: reactor volume = 5500 ft^3, waste density = 1 g/cm^3, radiation absorption = 100%.

(2) Dose rate from 1 Ci:

$$\frac{(3.7 \times 10^{10}\ \text{dis/sec}) \times (3.6 \times 10^{3}\ \text{sec/hr}) \times (2.5\ \text{MeV/dis}) \times (1.6 \times 10^{-6}\ \text{erg/MeV}) \times [1 \times 10^{-2}\ \text{rad/(erg)(g)}]}{(1\ \text{g/cm}^3) \times (2.83 \times 10^{4}\ \text{cm}^3\text{/ft}^3) \times (5.5 \times 10^{3}\ \text{ft}^3)} = 3.4 \times 10^{-2}\ \text{rads/hr}$$

(3) The dose per curie times the number of curies that can be afforded at allowable treatment costs (Fig. 3):

Allowable treatment cost, $/10^3 gal	^{60}Co ($0.35/Ci) dose, Mrads	^{60}Co ($0.10/Ci) dose, Mrads
0.20	0.017	0.058
0.50	0.068	0.238
0.80	0.119	0.408
1.00	0.153	0.526

value for G_{-O_2eq}, which must be increased to make radiation treatment economically feasible. The factors by which the G_{-O_2eq} values obtained in the laboratory must be multiplied to make radiation treatment economically feasible, based on a ^{60}Co cost of \$0.10/Ci, are:

Waste	Allowable treatment cost, $/10^3 gal	Multiplicative factor
2-Naphthalene sulfonic acid sodium salt	0.20	16
	0.50	5
	1.00	2
Chlorantine Fast Red	0.20	25
	0.50	7
	1.00	3

These values show that—depending on the waste considered and the allowable treatment costs—overall oxidation-reaction efficiencies presently obtainable must be increased by factors varying from 2 to 25 before radiation treatment will be economically feasible.

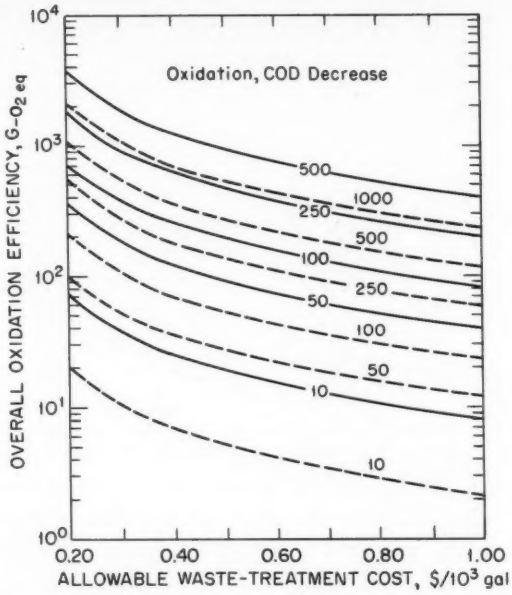

Fig. 4 Minimum G_{-O_2eq} values required for radiation-induced oxidation to be economically feasible. Cost: —, \$0.35/Ci; ---, \$0.10/Ci.

DISCUSSION

The increases in the overall oxidation-reaction efficiencies required to make radiation treatment of the two wastes economically feasible indicate that further work is needed. Although other wastes may provide a higher reaction yield using ionizing radiation as an energy source, the experimental results upon which this economic analysis is based are general enough for the conclusions drawn below. If the economic analysis had indicated that reaction rates needed were no more than twice those so far achieved, proponents of the use of radiation could feel encouraged. However, because the increases in reaction rates needed at the lower end of the allowable treatment-cost range (\$0.20/10^3 gal) are from 16 to 25 times those presently attainable, a major breakthrough is needed before radiation treatment of industrial wastes will become economically feasible. This barrier also

Table 3 Effect of Radiation on Real and Simulated Chemical Wastes*

Type of waste	Dose, Mrads	COD,† mgO_2/liter				5-Day BOD,† mgO_2/liter		COD/BOD	
		Before treatment	After treatment	Change	G_{-O_2eq}	Before treatment	After treatment	Before treatment	After treatment
2-Naphthalene sulfonic acid sodium salt (simulated waste)‡	1.0	305	165	140	18	0	40	∞	4.1
Chlorantine Fast Red (real dye waste)§	1.4	485	252	233	21	9	67	54	3.8

*Values are given for irradiation of samples under alkaline conditions (pH ≈ 11) and saturated with oxygen at 1 atm total pressure. These conditions give the best oxidation efficiency.

†COD is chemical oxygen demand; BOD is biological oxygen demand.

‡Data obtained at Aerochem Research Laboratories, Inc.

§Data taken from Ref. 3.

indicates the need for further work on understanding the basic chemistry involved. An investigation of the use of oxidation promoters, either soluble multi-oxidation-state cations or heterogeneous particles of transition elements and their oxides, may attract future effort. There is a body of literature on the subject of radiation catalysis,[4-6] but so far it has not been extensively applied to the treatment of industrial wastes.

CONCLUSIONS

Based upon the foregoing analysis, the conclusions reached are: (1) If radiation-induced oxidation pretreatment of industrial waste waters is to become economically feasible, the overall efficiency of the oxidation reactions must be increased by factors ranging from 2 to 25 times those presently obtainable. (2) For this large increase in reaction efficiency to occur, a better understanding of the chemistry involved will be necessary. (FEM)

REFERENCES

1. U. S. Department of Health, Education, and Welfare, Public Health Service, Advanced Waste Treatment Research Program, Summary Report for January 1962–June 1964, Publication Number 999-WP-24 (AWTR-14), April 1965.
2. U. S. Department of the Interior, Federal Water Pollution Control Administration, Advanced Waste Treatment Research Program, Summary Report for July 1964–July 1967, Publication WP-20-AWTR-19, 1968.
3. A. I. Mytelka and R. M. Manganelli, Energy-Induced Changes in an Azo Dyestuff Waste, *J. Water Pollut. Contr. Fed.*, 40(2): 260-268 (1968).
4. H. Haissinsky and M. Duflo, Heterogeneous Catalysis in Radiation Chemistry, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 29, pp. 47-51, United Nations, New York, 1958.
5. V. I. Veselovskii, N. B. Miller, and D. M. Shub, The Mechanism of Radiation Chemical Homogeneous and Heterogeneous Formation of Hydrogen Peroxide, *Sb. Rab. Radiatsionnoi Khim., Akad. Nauk SSSR*, pp. 49-60, (1955). [*Chem. Abstr.*, 50: 9115*g* (1955).]
6. M. Fleischman and R. H. Price, Gamma Irradiation of Dilute Aqueous Alkyl Benzene Sulfonate Solutions, *Environ. Sci. Technol.*, 1(7): 573-577 (1967).

Gamma Irradiation as a Pretreatment to Chemical Precipitation in the Purification of Domestic Sewage

By Lorne A. Campbell*

Supplementary Keywords: *source, gamma; pollution; water and waste treatment;* ^{60}Co.

Abstract: *Irradiation of raw sewage to a dose level of about 0.55 Mrad in 0.5 hr modified the carbonaceous impurities so that subsequent precipitation with lime increased carbon removal 10% over that effected by lime alone. The effluent was essentially free from phosphate and was sterile.*

The activated-sludge process followed by chlorination is widely considered to be the best process for the purification of municipal sewage. However, the process is biological and thus is difficult to control, and it does not completely remove phosphate. Prior to adoption of the activated-sludge process, lime was used to precipitate phosphate. Gamma irradiation of sewage prior to lime addition was considered a possible means of improving the lime-treatment process. The results of an experiment on such treatment are reported here.

PROCEDURE

Raw sewage from a small activated-sludge plant that treats domestic waste only was collected at the input to the plant and filtered through a coarse filter. The sewage was gamma irradiated in the ^{60}Co Gamma-cell 220 at a rate of 0.018 Mrad/min.

*Ontario Research Foundation, Sheridan Park, Ontario, Canada. This article is a summary of Canadian Report AM-6820.

XUM

RESULTS

Precipitation of Carbon-Containing Compounds and Phosphates

One-liter sewage samples (carbon, 68.5 mg/liter) were irradiated for different times and then treated with 600 mg of $Ca(OH)_2$ per liter. When the precipitate had settled, the supernatant was filtered through glass fiber, and the filtrate was analyzed for carbon. A portion of the filtrate was then treated with activated charcoal (0.4 g/liter), refiltered, and again analyzed for carbon. Doses up to about 0.3 Mrad decreased or did not affect the amount of carbonaceous material precipitated with lime, but 0.6 Mrad increased precipitation about 10%:

Irradiation time, min	Dose, Mrads	Carbon, mg/liter	
		After lime precipitation	After charcoal treatment
0	0	34.0	18.1
2	0.036	37.3	21.1
4	0.072	37.1	21.5
8	0.144	36.3	21.1
16	0.288	34.0	18.8
32	0.576	31.1	18.5

Carbonaceous compounds not precipitated by lime were also not adsorbed on activated carbon. The dose level was therefore standardized at 0.55 Mrad.

In further experiments, raw-sewage samples having a carbon content varying from 43.5 to 65.8 mg/liter were filtered through a Millipore HA filter (which removed all particulate matter larger than 0.45 μ) and were treated with $Ca(OH)_2$ (600 mg/liter) or with $Al_2(SO_4)_3 \cdot 18H_2O$ both as received and following a 0.5-Mrad gamma irradiation. The purification with respect to dissolved carbon averaged 17.1% with lime or alum alone but 26.2% when the the treatment was preceded by gamma irradiation. Aspirating the sewage with oxygen or nitrogen during the 30-min irradiation did not significantly change the results, nor did irradiating the mixture of sewage and lime.

The filtrate was essentially free from phosphate and, for all practical purposes, sterile. It was slightly alkaline with excess calcium hydroxide, which eventually forms calcium carbonate and precipitates.

The samples chosen for these experiments were from daytime hours. At this activated-sludge plant, the 24-hr average concentration of dissolved carbon in the raw sewage may be as low as 40 mg/liter, and treatment of 24-hr material should give a better effluent.

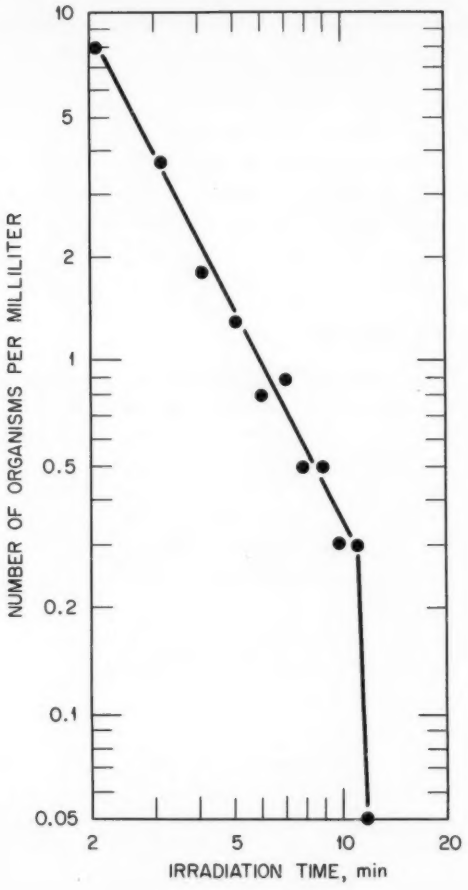

Fig. 1 Effect of gamma irradiation (0.018 Mrad/min) of raw sewage on survival of microorganisms.

Sterilization

Ten-milliliter samples of coarsely filtered raw sewage were irradiated and cultured on nutrient agar. There was a log-log relation (Fig. 1) between time and survivors (or kill) between 2 and 11 min (0.036 and 0.198 Mrad).

Gamma irradiation of lime-treated raw sewage rendered the filtrate sterile in less than 1 min, probably because most of the organisms had been removed in the lime slurry. (MG)

Gamma Irradiation of Waters and Waste Waters for Disinfection Purposes

By A. H. Vajdic*

Supplementary Keywords: *source, gamma; water and waste treatment; pollution; sterilization;* ^{60}Co.

Abstract: *Aqueous suspensions of* Escherichia coli *were sterilized by gamma irradiation much more rapidly in the presence of a low chlorine residual, but virus inactivation was not affected by up to 1.5 ppM chlorine. A chlorine residual of 1 ppM was decreased to a nondetectable level by irradiation with* 4.0×10^4 *rads. A gamma dose of* 3.5×10^4 *rads considerably decreased the threshold odor of water containing the taste and odor component produced by an actinomycete.*

Preliminary experiments to determine the applicability of gamma irradiation to disinfection of water and waste water containing residual chlorine and to the removal of substances responsible for taste and odor are described. The combined effect of gamma radiation and chlorine in water sterilization was studied earlier by Sidorenko, Pinigin, and Savenkova.[1]

For the disinfection studies, the bacterium *Escherichia coli* and a bacterial virus, a bacteriophage of *E. coli* B, were used. The gamma source was the ^{60}Co Gammacell 220, which provides an irradiation rate of about 450 thousand rads/hr.

DISINFECTION

Bacteria

Suspensions of 18-hr cultures of *E. coli* (grown at 35°C in nutrient broth) were made up in tap water to contain ~10^8 cells per 100 ml. Irradiation with a dose of 3.4×10^4 rads (about 5 min exposure) in the presence of 0.35 ppM chlorine killed 100% of the organisms. In the absence of chlorine, 10^5 organisms per 100 ml remained, while with chlorination alone over the same time period, 10^4 remained.

In the determination of the chlorine residual remaining in the suspensions after 5 min, none was found in the chlorinated suspension exposed to 3.4×10^4 rads. Tests made with 1 ppM chlorine indicated that this amount would be decreased to nondetectable levels by 4.08×10^4 rads.

Virus

Aqueous suspensions of the bacteriophage of *E. coli* B containing about 10^8 particles per 100 ml were treated similarly, except that the chlorine residual was 1.5 ppM. The results were quite different from those with *E. coli* since the inactivation achieved with the combined treatment was little different from that caused by irradiation alone.

Virus survivors were enumerated by the most probable number method,[2] which is particularly applicable where low numbers pertain.

TASTE AND ODOR REMOVAL

Water with a taste and odor typical of that produced by actinomycete species was prepared by adding a methylene chloride extract[3] of an actinomycete (streptomyces species) culture, exhibiting the typical earthy odor, to a large volume of distilled water. Aliquots of this water had an initial threshold odor of more than 600. A radiation dose of 3.5×10^4 rads decreased the threshold odor to 17, and 3.5×10^5 rads decreased it to 12. The value of 17 is comparable to that obtained by contact with 50 ppM activated carbon for 30 min. (MG)

REFERENCES

1. G. L. Sidorenko, M. A. Pinigin, and V. T. Savenkova, The Use of the Combined Effect of Gamma Irradiation and Chlorine for the Sterilizing of Water (in Russian), *Gig. Sanit.*, 28(9): 97-98 (1963) (English translation in Report JPRS-21,915, pp. 10-12).
2. A. H. Vajdic, The Enumeration of Bacteriophages in Water and Sewage Using the Most Probable Number Method, Ontario Water Resources Commission, Research Publication No. 26, 1967.
3. N. N. Gerber and H. A. Lechevalier, Geosmin, an Earthy-Smelling Substance Isolated from Actinomycetes, *Appl. Microbiol.*, 13: 935-938 (1965).

*Division of Research, Ontario Water Resources Commission.

XUM

Gamma Irradiation of Raw Sewage for Sedimentation Purposes

By R. H. Andrews and M. B. Fielding*

Supplementary Keywords: *source, gamma; facility description; water and waste treatment; pollution; precipitation;* ^{60}Co.

Abstract: *In preliminary tests, gamma (^{60}Co) irradiation of raw sewage did not consistently lead to improved settling; solids degradation decreased the settling.*

With increased emphasis being placed on advanced waste-treatment processes and with the necessity of improving existing processes, the Division of Research of the Ontario Water Resources Commission and the Commercial Products Division of Atomic Energy of Canada Limited are cooperating in an investigation of the use of gamma irradiation as a practical aid to sewage treatment. The work discussed here was a very preliminary attempt to determine the gross results of gamma irradiation rather than causes and effects. For this reason, actual raw sewages were used, despite their complexity and lack of reproducibility.

PROCEDURE

The irradiation facilities used in the studies were two Gammacells, Models 220 and 200 (loaned and installed by Atomic Energy of Canada Limited), loaded with 4800 and 236 Ci of ^{60}Co, respectively. Samples up to 8 in. in height and 6 in. in diameter (in the 220) may be irradiated. Within the lead shielding of the facility, a cylindrical carrier, with a chamber in which samples may be placed from outside the shield, may be raised or lowered. A digital timer, preset to give the desired exposure, automatically raises the carrier at the end of the irradiation. Average dose rates within the sample chambers of Models 220 and 200 averaged about 7100 and 630 rads/min, respectively, over the test period. Irradiation times were adjusted every 2 weeks to correct for source decay. Samples of 1 liter volume could be processed in Model 220 and of 500 ml in Model 200.

Raw sewages from a water-pollution-control plant containing both industrial and domestic wastes and from a residential area containing largely domestic sewages were used. The effects of irradiation on the settling of the sewages at room temperature were determined, along with biological oxygen demand (BOD), chemical oxygen demand (COD), total solids, suspended solids, and dissolved solids.

The sewages were irradiated in glass beakers with total doses of 10^2 to 10^6 rads (Gammacell 200) and with 10^3 to 10^7 rads (Gammacell 220). The usual procedure was to collect enough sewage to last 1 day, and, after screening off large solids with a 0.141-cm (0.0555-in.) mesh sieve, irradiate the sewage, and then allow a 60-min settling in Imhoff cones. Temperature rises of the sewage during irradiation were less than 2°C except when exposure times were longer than 1 hr in the Model 220.

RESULTS

Settling of irradiated samples was expressed as a percentage of that in a control sample, with positive results indicating improved settling. In general, settling of solids from domestic sewage was increased about 20% by a total radiation dose of 10^6 rads but then was decreased sharply by higher doses (Fig. 1), indicating degradation of the solids. In mixed sewage, improvement was only about 10% with this dose, and this also was followed by a short drop. Results were similar with the 630 rad/hr dose rate, the degradation occurring at about 10^5 rads.

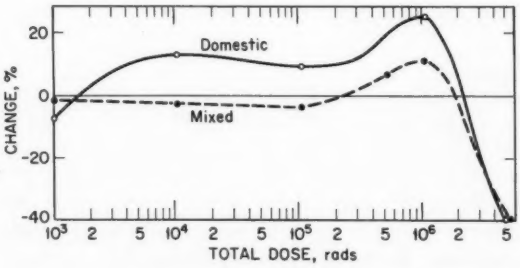

Fig. 1 Effect of gamma (^{60}Co) irradiation on settling of solids from raw sewage irradiated at a rate of 7100 rads/min.

*Division of Research, Ontario Water Resources Commission.

On days when the raw sewage was high in BOD and suspended solids, the volume of settled solids was high, but the percent changes after irradiation were low. There were not enough of these days to statistically correlate high solids with changes in settling, but there did not appear to be any differences in settling characteristics from those observed on average days.

The day-to-day results with sewage collected from the same source at the same time of day differed considerably, which reflects the variable composition of the sewages. However, values were consistent for several samples collected at the same time on any one day and subjected to the same total dose. Some scatter is accounted for by the small volume—1 to 2 ml— of settled solids found after 60 min since a difference of only 0.3 ml can correspond to a 30% change.

Chemical analyses showed little alteration in the irradiated samples except at doses of 10^6 rads or more. At high total doses, BODs and CODs of settled irradiated samples (taken from the supernatant) were slightly but consistently lower than their settled controls, indicating that irradiation had contributed to oxygenation of the samples.

(MG)

Destruction of Organic Substances in Waste Water by Ionizing Radiation*

By D. M. J. Comptont

Supplementary Keywords: *source, gamma; water and waste treatment; pollution; economics.*

Abstract: *The results of a study of direct radiation-induced oxidation as a means of decreasing the chemical oxygen demand (COD) of waste waters indicated that the technique is not yet economically competitive with other processes. Typical changes in COD were found to be 8 to 10 mg/liter for radiation doses of 10^5 rads delivered with aeration, at a cost of 50 cents to $1.00 per 10^3 gal for the irradiation.*

Dissolved organic impurities are usually removed from waste waters by biological oxidation in the activated-sludge process or in the trickling-filter process. The fractional removal that is possible with biological treatment is limited by the fairly high frequency of failure of the process due to toxicity or sudden load changes and to the reaching of a steady state in which organic products of cell decomposition are released into the water as fast as organic materials are removed. These limitations have led to an intensive search for methods of treating sewage which might offer improved performance at a lower cost.

One such technique is the use of ionizing radiation. This has been studied by Etzel's group at Purdue University, who have reported[1] a radiation-induced decrease in chemical oxygen demand in synthetic waste water and in glucose and phenol solutions.[2] A thesis by Alexander[3] treats changes in biodegradability produced by irradiation. Fleischman and Price[4] have reported the destruction of alkylbenzene sulfonate (ABS), and Cohen[5] includes in a thesis a study of the degradation of ABS and emphasizes that aeration of the water is necessary. The use of ionizing radiation to oxidize organic substances in solution has also been investigated by Gulf General Atomic under contract to the AEC Division of Isotopes Development. Data on the oxidation and removal of organic impurities in waste waters by use of the gamma radiation from spent fuel elements, using actual waste waters, and a description of the radiation source used are reported here together with a theoretical model and the results expressed in engineering units capable of economic analysis. Portions of this work have been reported elsewhere.[6]

THEORETICAL CONSIDERATIONS

Waste waters are characterized by a number of parameters. Their oxygen demand (OD) is the amount of oxygen required to oxidize the oxidizable im-

*Summary of Report GA-9821, paper presented at the 62nd Annual Meeting of American Institute of Chemical Engineers, Washington, D. C., November 1969.

†Gulf General Atomic Incorporated, P. O. Box 608, San Diego, Calif. 92112; present address: Enviro-Med, Inc., La Jolla, Calif.

XUM

purities, and the chemical oxygen demand (COD) and biological oxygen demand (BOD) are the ODs measured using chemical and biological oxidants, respectively. The total organic carbon (TOC) is determined by oxidizing the organic carbon to an equivalent amount of carbon dioxide, which is measured. Inorganic carbon is distinguished from organic by acidification. Typical primary effluents (the supernatant flow from the primary sedimentation process) may have COD values from 60 to 300 mg/liter. Secondary (biological) treatment may decrease the COD of the secondary effluent to 20 to 40 mg/liter. The G value is the number of molecules changed (in this case, single-electron oxidations) per 100 eV of ionization energy deposited. The radiation dose, expressed in rads, can be converted to electron volts per liter in water by the relation 1 rad = 100 ergs/g = 6.2×10^{16} eV/liter.

If we take a G value of G_{ox}, a radiation dose of R rads, and an oxidation equivalent weight for oxygen of 8, the change in COD (ΔCOD) produced will be approximately $8 \times 10^{-6}\, RG_{ox}$. For example, with $G_{ox} = 10$ and $R = 10^5$ rads, the ΔCOD would be 8 mg/liter. The cost of delivering a dose of 10^5 rads to waste water is estimated by Ballantine (article in this issue of *Isotopes and Radiation Technology*) as possibly about 83 cents per 10^3 gal. By contrast, the *total* cost of sewage treatment using a conventional activated-sludge process removing 90% of the COD of the primary effluent is less than 20 cents per 10^3 gal. High G_{ox} values would thus be needed for an economically competitive process.

The G values that are likely to be observed can be estimated by considering the radiation chemistry involved. When water is irradiated, the chief primary species formed are the hydrated electron, e^-_{aq}; the hydrogen atom, $H^\cdot$; and the hydroxyl radical, $OH^\cdot$. The G values for formation are about 2.6, 0.5, and 2.6, respectively. These species may react with each other or with impurities. In pure neutral water only the OH radical is an oxidant, and a G_{ox} near 3 would be expected. If H_2O_2 has been added or has built up, the reaction $e^-_{aq} + H_2O_2 \rightarrow OH + OH^-$ can occur, so that a G_{ox} near 6 might be obtained. In water containing oxygen, the oxidizing power of the dissolved oxygen (DO), which itself is incapable of oxidizing most organic materials, can be made available by reactions of e^-_{aq} and $H^\cdot$ to give O_2^-, which is a powerful oxiding species, or HO_2, which dissociates in a nonacid medium to give O_2^-. A further increase in the G_{ox} value can be obtained only if a chain reaction can be established in which further dissolved oxygen can be made available for oxidation. Chain reactions of this type have been observed by Broszkiewicz,[7] Stolarczyk,[8] and Dewhurst,[9] who studied the effects of organic compounds on the radiation oxidation of Fe^{2+} and found G_{ox} values typically less than 50. Similar short-chain reactions have also been observed[10] in catalytic oxidation of waste waters by $H_2O_2 + Fe^{2+}$ and in the study[11] of radiation treatment of acid mine wastes.

These considerations indicate that the G value for oxidation of organic substances present in waste water will be higher in aerated solutions than in nonaerated—e.g., 10 to 15 or more vs. 3 to 5. Since the DO content of water in equilibrium with air at atmospheric pressure is approximately 8 mg/liter, this enhancement can be maintained only until the oxygen originally dissolved is consumed, unless the aeration is continued. If the radiation dose is delivered at a high dose rate, e.g., from an accelerator, so that the DO is consumed faster than it can be replenished by aeration, the oxidation will be similar to that under nonaerated conditions.

When excess oxidizable organic material is present, the COD should decrease linearly with decreasing radiation dose and independently of the COD value. This linear decrease should continue until the COD is low enough for processes other than oxidation to become significant. Accordingly, a statement that destruction of 90 or 95% of an organic material dissolved in water occurs with a certain radiation dose is not significant unless the initial concentration is specified. Plainly, the fraction destroyed will increase as the initial concentration decreases. Furthermore, measurements of the destruction of an organic material dissolved in pure water may differ greatly from those made on the same material dissolved in waste water, where the material under consideration must compete with other organic materials for the active species. For some pure substances, the results of competition can be predicted from rate-constant tables.[12]

Since the greater part of the oxidation is due to the DO made available by free-radical attack, the G value for oxygen uptake, G_{-O_2}, is a convenient measurement to make. This value can give information about chain reactions and about possible competition between dissolved substances and DO for the primary products of the irradiation. For a nonchain reaction, in which DO is attacked by e^-_{aq}, the G_{-O_2} value would be expected to be about 2.6. Measurements of G_{-O_2} are more sensitive than those of COD and also provide a means of formulating an oxidation balance. If all the oxidation should be due to activation of the dissolved

XUM

O_2 to O_2^- and each O_2^- should yield three equivalents of oxidation produced by the OH formed when $G_{OH} = 2.6 = G_{-O_2}$, then $G_{ox} = 4G_{-O_2}$ and $\Delta COD = \Delta DO$.

RESULTS

Theoretical predictions were borne out by the results of our experiments. From a typical measurement of DO uptake (Fig. 1) during gamma irradiation (using spent fuel elements), the DO uptake in distilled water is found to be about 10.5 mg/liter per 100 krads, giving a G_{-O_2} value of approximately 3.3. This corresponds to an attack on O_2^- by e_{aq}^- and $H^\cdot$ with no chain reaction. The G_{ox} value (for four equivalents of oxidation per O_2 molecule taken up) is approximately 13. With primary sewage-plant effluent, the uptake is 12.8 mg/liter per 100 krads, indicating that oxygen can compete effectively with organic impurities for the primary irradiation products. The higher value may be due to the suppression of the back reaction, $HO_2 + H^\cdot \rightarrow H_2O + O_2$, by the capture of HO_2 by organic substances. Similar values have recently been reported by Evans[13] for water and water containing bacteria. That the value is only slightly higher indicates that no significant chain reactions are occurring. The DO uptake by secondary effluent is intermediate in value. Results were similar with more than 50 samples of effluents, and in no case was the observed G_{-O_2} value higher than 3.5, even when possible catalysts such as NO_3^-, Fe^{2+}, and H_2O_2 were added.

The DO uptake and COD decrease were measured on samples of primary effluent that were completely deaerated by sparging with nitrogen, presaturated with air or oxygen but with no replacement of oxygen, or continuously aerated or oxygenated with air or oxygen at atmospheric pressure. In some cases agreement between the two measurements was satisfactory (Fig. 2*a* and *b*). For example, with a sample of primary effluent, the uptake was about 8 mg/liter per 10^5 rads, corresponding to a G_{-O_2} of 2.5. The COD of another sample of the same material irradiated during continuous aeration decreased initially at a rate of 8.5 mg/liter per 10^5 rads, corresponding to a G_{ox} of 10. At low COD values (less than 80), the ΔCOD decreased, as shown by the last point on Fig. 2*b*. This behavior is typical. That the irradiated effluent contains substances capable of competing with DO for the primary products of the irradiation is indicated by the DO uptake of 5.9 mg/liter per 100 krads for an irradiated sample of primary effluent previously irradiated, during aeration, with 3.3×10^6 rads (Fig. 3). There was no decrease in COD in a nonaerated control.

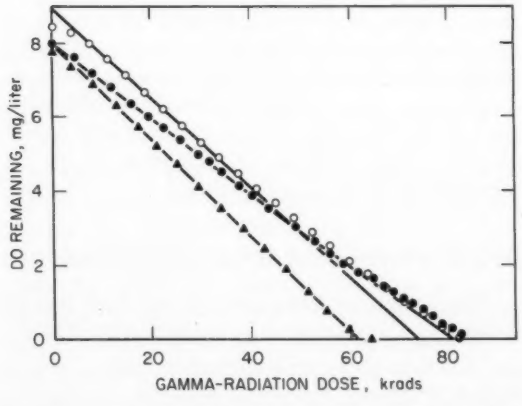

Fig. 1 Decrease in DO uptake in (▲) distilled water, (●) primary sewage-plant effluent, and (○) secondary sewage-plant effluent during gamma irradiation.

In many cases the COD decrease was less than that shown in Fig. 2*b*, but in no case was it greater. Continuous aeration is necessary to achieve the maximum rate of decrease of COD, and nonaerated samples did not show G_{ox} values greater than 3. Bubbling with oxygen gas during irradiation did not give any greater decrease in COD than when air was used, indicating that a DO level of 8 mg/liter is sufficient to capture all the primary products of irradiation that can be captured. Measurements of TOC showed decreases typically less than half the COD decreases, indicating that less than half the oxidation produces CO_2 and H_2O, the other half producing more highly oxidized forms, e.g., acetic acid from ethanol, which do not register as a change in TOC.

We thus conclude that neither COD nor DO measurements give evidence of chain reactions or high G values. This conclusion is supported in detail by analysis, in the same terms, of the work of other authors, e.g., Condren[14] and Touhill *et al.*[15] Condren's work, for example, shows a decrease in COD of phenol solutions which are initially linear with radiation-dose change and has a slope corresponding to $\Delta COD \approx 8$ mg/liter per 100 krads. The DO uptake of 12.8 mg/liter per 100 krads by a solution of 100 mg/liter of phenol in water (Fig. 3) indicates that phenol does not compete effectively with dissolved oxygen for e_{aq}^- and $H^\cdot$ but is effectively oxidized by $OH^\cdot$, HO_2, and O_2^-.

Since there are no signs of high G values or of chain reactions, and since these results are in good agreement with a simple theory, we may safely conclude that

XUM

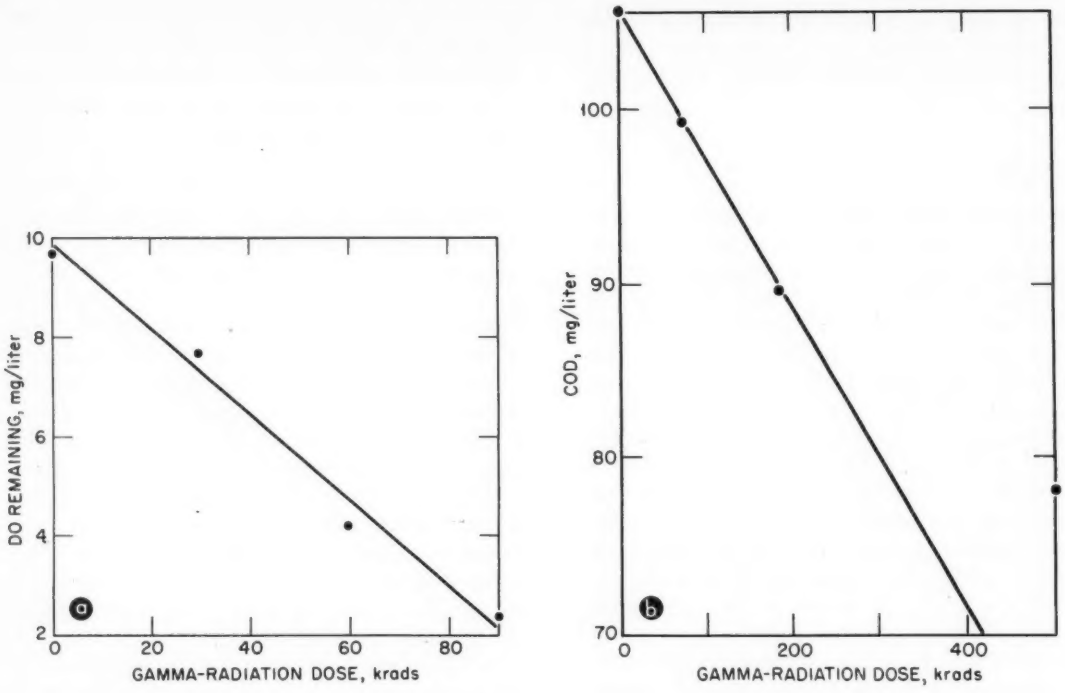

Fig. 2 (*a*) **DO remaining in a sample of primary effluent and** (*b*) **COD of another sample of the same material as a function of gamma-radiation dose, at a dose rate of 3×10^5 rads/hr. In each case, successive measurements were made on a single sample.**

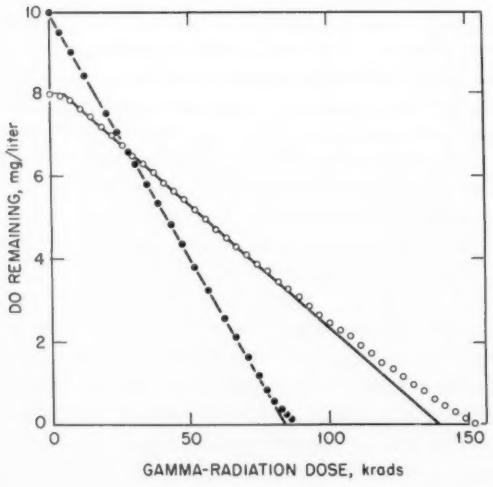

Fig. 3 Effect of dose on DO for (○) primary effluent that had been irradiated with 3.3×10^6 rads during aeration and (●) a solution of 100 mg/liter of phenol in distilled water. Measurements were made during gamma irradiation.

reduction of COD by simple irradiation is not an economically competitive process, either as a replacement for secondary treatment or as a tertiary treatment. A dose of 10^5 rads will decrease the COD by only 8 to 12 units, and the decrease in COD tends to reach a limit with increasing dose. Treatment of municipal waste waters by direct radiation oxidation thus does not appear economically attractive compared to processes such as carbon adsorption, which also removes dissolved organic impurities, or to processes such as reverse osmosis or distillation, which can potentially produce water fit for reuse.

DESCRIPTION OF RADIATION SOURCE*

The gamma radiation used in the Gulf General Atomic tests was supplied by mixed fission products.

*Added from Report GA-9924 by W. L. Whittemore, D. M. J. Compton, and S. J. Black, Ionizing Radiation for the Treatment of Municipal Waste Waters, Final Report, Feb. 26, 1969–Feb. 28, 1970.

XUM

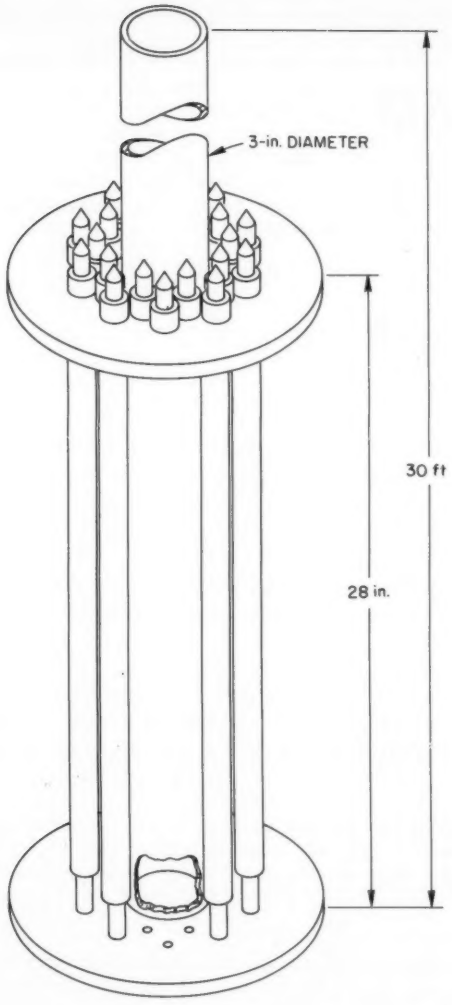

Fig. 4 Arrangement of 19 spent TRIGA fuel elements concentrically around dry irradiator tube.

For static tests, 19 spent fuel elements from the TRIGA reactor were arranged concentrically (Fig. 4) around the irradiation chamber, which could accommodate samples of up to 1 liter. Within the 3-in.-diameter cylindrical chamber, the dose rate as a function of radius was uniform, although there was, of course, a vertical variation of radiation intensity. A larger number of fuel elements would have increased the dose rate but could not be used because of the type of license in effect. However, with more elements, the radiation efficiency would have been less because two concentric rings would have been required.

On-line waste-water treatment was tested under conditions simulating those that would obtain in an actual sewage plant. The waters were circulated in a system consisting of multiple passes of tubing located in the gamma field of a large TRIGA reactor. First, the reactor, at a distance from the tubing unit, was operated at 1.5 MW for 0.5 hr. The reactor was then shut down and, after a 10-min interval to allow delayed neutrons to die away, the reactor was moved close to the tubing unit so that the water circulating in the tubes was exposed to the gamma radiation from the reactor. (MG)

REFERENCES

1. A. J. Condron [Condren] and J. E. Etzel, Destruction of Dilute Organic Pollutants by Gamma Irradiation, *Effluent Water Treat. J.*, 7: 528-532 (1967).
2. J. E. Etzel and A. J. Condren, Radiation Treatment of Wastewaters, *Trans. Amer. Nucl. Soc.*, 11: 56-57 (1968).
3. M. L. Alexander, Biodegradability of Sewage Following Gamma Irradiation, Thesis, Purdue University, Lafayette, Indiana, 1969.
4. M. Fleischman and R. H. Price, Gamma Irradiation of Dilute Aqueous Alkyl Benzene Sulfonate Solutions, *Environ. Sci. Technol.*, 1: 573-577 (1967).
5. J. B. Cohen, Effects of Ionizing Radiation on Aqueous Solutions of the Detergent Alkylbenzene Sulfonate and Several Lower Homologs, Thesis, California Institute of Technology, Pasadena, Calif., 1965 (*Dissertation Abstr.*, 26: 85-86).
6. D. M. J. Compton, S. J. Black, F. L. Lieurance, and W. L. Whittemore, Application of Ionizing Radiation to the Treatment of Waste Waters and Sewage Sludge, in *Large Radiation Sources for Industrial Processes*, Symposium Proceedings, Munich, Aug. 18–22, 1969, pp. 399-416, International Atomic Energy Agency, Vienna (STI/PUB/236) (Gulf General Atomic Report GA-9632; CONF-690805).
7. R. Broszkiewicz, Transformations of Ferrous and Ferric Ions in Aqueous Solutions Directly Observed During Irradiation with Gamma Rays, in *Radiation Chemistry*, Symposium Proceedings, Tihany, 1962, Hungarian Academy of Sciences, Budapest, pp. 333-336 (1964).
8. L. Stolarczyk, Post-Effects of Gamma- and Electron-Pulse Irradiation on Aqueous Solutions of $FeSO_4$, H_2SO_4 and C_2H_5OH, *ibid.*, pp. 395-403.
9. H. A. Dewhurst, Effect of Aliphatic Alcohols on the Gamma-Ray Oxidation of Aerated Aqueous Ferrous Sulphate, *Trans. Faraday Soc.*, 48: 905-911 (1952).
10. D. F. Bishop, G. Stern, M. Fleischman, and L. S. Marshall, Hydrogen Peroxide Catalytic Oxidation of Refractory Organics in Municipal Waste Waters, *Amer. Chem. Soc., Div. Water, Air, Waste Chem., General Papers*, 4: 85-90 (1964); and *Ind. Eng. Chem., Process Design Develop.*, 7: 110-117 (1968).

XUM

11. M. Steinberg, J. Pruzansky, L. R. Jefferson, and B. Manowitz, Removal of Iron from Mine Drainage Waste with the Aid of High Energy Radiation, USAEC Report BNL-11576, Brookhaven National Laboratory, 1967.
12. M. Anbar and P. Neta, A Compilation of Specific Bimolecular Rate Constants for the Reactions of Hydrated Electrons, Hydrogen Atoms, and Hydroxyl Radicals with Inorganic and Organic Compounds in Aqueous Solution, *Int. J. Appl. Radiat. Isotop.*, **18**: 493-523 (1967).
13. N. T. S. Evans, Removal of Dissolved Oxygen from Aqueous Media by Ionizing Radiations, *Radiat. Eff.*, **1**: 19-22 (1969).
14. A. J. Condren, Radiation-Induced Oxidation of Selected Organics in Waste Water, Thesis, Purdue University, Lafayette, Indiana, 1969 (*Nucl. Sci. Abstr.*, **24**: 33722).
15. C. J. Touhill, E. C. Martin, M. P. Fujihara, J. E. Stein, and G. McDonnell, The Effects of Radiation on Chicago Metropolitan Sanitary District Municipal and Industrial Wastewaters, *J. Water Pollut. Contr. Fed.*, **41**(Part 2): R44-60 (February 1969).

Section

III

Isotopes and Radiation Technology

Miscellaneous

Recent Reports on Pesticides

Pesticide-residue problems are becoming increasingly important because, on the one hand, the rapid world-population increase demands greater use of pesticides while, on the other, public sensitivity to contamination of food, wildlife, and the total environment is growing. Three recent reports on pesticide problems will be of interest to the environmental scientist:

Nuclear Techniques for Studying Pesticide Residue Problems, Panel Proceedings, Vienna, Dec. 16–20, 1968 (FAO/IAEA), IAEA, Vienna, 1970 (STI/PUB/252); 104 references; $3.00.

Supplementary Keywords: *chemistry, analytical; biochemistry; botany; ecology; agriculture; industry, food; material tracing; preparation, labeled material; activation, neutron; safety; bibliography; textbook; pollution;* ^{3}H; ^{14}C; ^{32}P; ^{35}S; ^{36}Cl; ^{82}Br; ^{203}Hg.

Isotope tracer and radioactivation techniques are powerful and unique tools for selecting pesticides and establishing conditions for their safe and effective use. To encourage exploitation of nuclear techniques in solving problems and to advise the Joint Food and Agriculture Organization/International Atomic Energy Agency (FAO/IAEA) Division on its program relative to the problems, a panel of 19 participants from 10 countries met at the IAEA headquarters. This publication contains the eight papers presented and the report and recommendations of the panel. The papers discuss the food-chain persistence and the metabolism—as determined by nuclear methods—of the residues of commonly used pesticides. Particular attention is given to thion and to the organochlorine, organobromine, and organomercurial compounds. Also, the toxicity of pesticides labeled with radionuclides is considered. International cooperative activities on the problems of pesticide residues in food are outlined.

The panel report discusses the need for integrated experiments, the value of tracer techniques, problems that require solution, and suggested pilot studies. Recommendations include an internationally coordinated research program, administrative support of the program, improved facilities for the use of isotope tracers in studying the fate of pesticide residues, and the assembly and dissemination of information on nuclear methods in pesticide residue study. (HPR)

Radioisotopes in Pesticide Research. IV, V (in German), *Isotopenpraxis,* **6**: 204-214 (1970).

The literature from 1967 through 1969 (631 references) related to the use of radioisotopes in pesticide research is reviewed. Three earlier reviews in *Atompraxis* [**10**: 65 (1964); **13**: 126, 202 (1967)] constituted parts I, II, and III. Even persons who do not read German will find this article useful because of the bibliography. (MG)

Regional Variation in Percutaneous Penetration in Man: Pesticides, by H. Maibach, R. Feldmann, T. Milby, and W. Serat, paper presented at American Academy of Dermatology meeting, Boca Raton, Fla., 1970.

A ^{14}C-labeled pesticide was used to show that the skin of the forearm is less penetrable to this chemical than that of the forehead, the angle of the jaw, back of the ear, the scalp, and the scrotum. The tests consisted in applying the chemical to the washed skin of volunteers, allowing it to remain for 24 hr, and determining the labeled compound in the urine excreted during this period. (MG)

XUM

Pollution Problems

^{197}Hg USED TO DETERMINE MERCURY IN FISH*

Supplementary Keywords: *activation, neutron; activation analysis; reactor; chemistry, analytical; ecology; method; ion exchange; comparison; pollution; mercury.*

Mercury in fish at concentrations as low as ~3 ppB was determined by neutron activation analysis followed by sorption of the ^{197}Hg formed—complexed as $HgCl_4^{2-}$—on Dowex 2 x 8 ion-exchange resin and counting directly on the resin, thus eliminating chemical steps that cause losses and irreproducibility. By gamma spectrometry, the area under the ^{197}Hg 77.3-keV photopeak is measured and is used to calculate the amount of mercury in the sample. The results compare favorably with those obtained on the same samples in other laboratories by several methods (atomic absorption, X-ray fluorescence, destructive and nondestructive NAA) and have a smaller relative standard deviation (±10% for mercury concentrations in the range 0.05 to 10 ppM).

Analyses made with the method showed the mercury content of most of the organs—the liver excepted—of a Lake Erie whole Coho salmon to be less than that of its edible tissue. The analysis of fish from several bodies of water remote from sources of mercury pollution showed that mercury-free fish-tissue samples ("zero" blanks) are possibly not obtainable.

(HPR)

NUCLEAR METHOD DETECTS 20 ELEMENTS IN PARTICULATES FROM CHICAGO AIR†

Supplementary Keywords: *activation, neutron; reactor; chemistry, analytical; spectrometry; statistics; pollution; atmosphere.*

Neutron activation followed by gamma-ray spectrometry detected 20 elements in air particulate samples collected on cellulose filters simultaneously at 22 surface locations in the Chicago metropolitan area. A 1-cm^2 portion of each filter was irradiated with thermal neutrons in the CP-5 research reactor at the Argonne National Laboratory, and induced nuclides were measured by gamma-ray spectrometry with NaI(Tl) crystals and a 400-channel analyzer. The detected elements were, in order of decreasing concentration (2400 to 0.12 ng/m^3): Fe, Cl, Al, Zn, Mn, Na, Br, V, Cr, Sb, Hg, Se, Ce, Ag, Co, La, Sc, Cs, and Eu; Ca was not measured quantitatively.

Least-squares computations of correlations between all possible pairs of elements and between elements and dust showed, for a particular level of significance, a linear correlation when the corresponding correlation coefficient was equal to or greater than a given value. Correlations of elements with dust and with one another indicate either that the elements had a common origin or that large volumes of airborne dust approached uniform mixing before the air passed over the filters.

No correlation was shown for vanadium and mercury with cobalt, with dust, with other elements, or with one another.

Concentrations of vanadium, the major source of which is fuel oil used for space heating, were highest in areas of highest population density. The sources of mercury and the reason for its noncorrelation are not known. The limited correlation of cobalt may have been due to the presence of cobalt in the general airborne dust or in localized sources.

The method is suggested as a possible way to inventory global surface air for some of its trace elements. (HPR)

COMPUTER PROGRAM HELPS DESIGN STACKS FOR CURBING AIR POLLUTION‡

Equations are given for determining the effective stack height from plume rise due to momentum and to buoyancy, diffusion of exiting gases, and deposition of particles. The input data are scanned by the computer,

*From J. M. Rottschafer, J. D. Jones, and H. B. Mark, Jr., A Simple Rapid Method for the Determination of Trace Mercury in Fish via Neutron Activation Analysis, presented at the 160th meeting of the American Chemical Society, Chicago, Ill., September 1970.

†From S. S. Brar, D. M. Nelson, J. R. Kline, P. F. Gustafson, E. L. Kanabrocki, C. E. Moore, and D. M. Hattori, Instrumental Analysis for Trace Elements Present in Chicago Area Surface Air, *Journal of Geophysical Research*, 75(15): 2939-2945 (1970).

‡Summary of article by D. L. Robins and M. M. Mattia (Day & Zimmermann, Inc.), *Chemical Engineering*, 75(3): 119-122 (Jan. 29, 1968).

XUM

which then operates on the stack-gas dispersion programs if the particulates are $<20\,\mu$ or the stack-particulate dispersion program if the particulates are $>20\,\mu$. The computations are made in matrix fashion.

With the computer program, the engineer can evaluate potential pollution from either gases or particulates and thus design economical and safe stacks. (MG)

Availability of Isotopes and Services

PORTABLE IRRADIATOR WITH ^{90}Sr SOURCE

The Model C irradiator offered for sale by the Palo Alto Laboratories of Teledyne-Isotopes (4062 Fabian St., Palo Alto, Calif. 94303) is designed for use where a high dose rate is needed for a small sample. The outside dimensions of its sealed aluminum housing are 6 by 6 by 6 in., and its total weight is 20 lb. The removable sample holder is a 14.5-mm-diameter by 9.0-mm-high polyethylene cup. The unit has a complete interlock system, which prevents radiation leakage during sample loading.

There are two ^{90}Sr–^{90}Y (28-year half-life) sources in an opposed geometry (Fig. 1). The ^{90}Y emits a 0.94-MeV (average) beta particle. The dose rate is around 240 rads/min from a 100-mCi source, and the internal dose rate is uniform to ±5% across a 14.5-mm-diameter object. The external rate is <0.03 mrem/hr per millicurie of total activity at 1 ft from the irradiator surface.

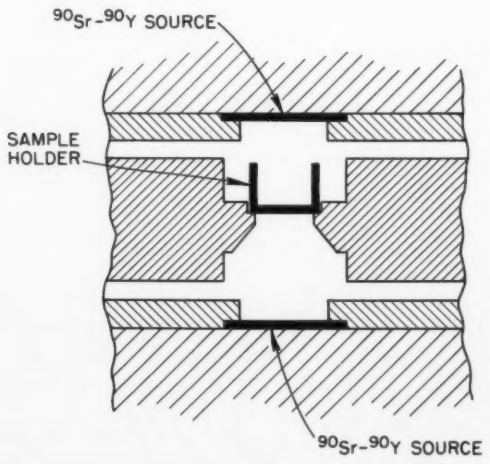

Fig. 1 Beta irradiator with ^{90}Sr–^{90}Y source.

The cost varies from $850 for the 2-mCi unit to $1195 for the 100-mCi unit.

NEUTRON ACTIVATION ANALYSIS FOR TRACE-MERCURY DETERMINATION

Atomic Energy of Canada Limited (AECL) has been analyzing mercury-containing samples for government health departments as well as for certain other institutions (e.g., hospitals) and is now offering a full-fledged service to manufacturers, public authorities, and researchers. Mercury in concentrations of 10^{-9} g can be determined. Other elements (60 in all) can be detected at the same time, though the high flux creates some interference problems unless a lithium-drifted germanium spectrometer is used instead of the conventional sodium iodide scintillation crystals. AECL is building a 2-kW prototype reactor that could be installed at reasonable cost in any laboratory complex. The prototype, to be set up at AECL's Ottawa laboratory, is an engineered version of an operating Slowpoke (Safe Low Power Critical Experiment) at Chalk River Nuclear Laboratories, which uses 1.5 lb of ^{235}U immersed in water and is surrounded by a beryllium reflector.

^{133}Xe INJECTION IN DISPOSABLE SYRINGES

The Radiochemical Centre in Amersham (Buckinghamshire, England) is supplying ^{133}Xe syringe units for injection. The product gives the user 10 mCi of ^{133}Xe in 1 ml of saline solution in a form that can be injected into a patient as a single dose. Gas bubbles are completely absent from the cartridge. The product (Code XAS.21P) is dispatched each Friday and assayed as of the following Monday; orders should be received by the Tuesday preceding dispatch. Inquiries are invited for syringes delivering quantities other than 10 mCi.

UNION CARBIDE PRODUCING FISSION PRODUCT ^{99}Mo

Union Carbide Corporation (P. O. Box 324, Tuxedo, N. Y. 10987) is producing ^{99}Mo by bombardment of highly enriched ^{235}U. The product, with a specific activity of 10 thousand Ci/g, will sell for 22.5 cents per millicurie, compared with a current price of 12 cents per millicurie for material with a specific activity of only 1 Ci/g.

ORNL ENRICHED-STABLE-ISOTOPE INVENTORY

Additions to the Oak Ridge National Laboratory (ORNL) enriched-stable-isotope saleable inventory through Dec. 31, 1970, include the following:

Isotope	Purity, %	Amount available, g
^{17}O	10.3	1.6
^{112}Sn	83.6	4.8
^{122}Te	96.2	2.0
^{164}Er	73.6	1.0
	67.2	1.4

Planned additions to the inventory during the next 6 months:

Isotope	Lot No.	Estimated recovery, g	Estimated mass assay, at.%
^{123}Sb	NN 164601	35	98
^{125}Te	NS 166201	46	95
^{126}Te	NS 166301	112	98
^{112}Sn	OT 178404	10	82
^{114}Sn	OT 178501	17	60
^{115}Sn	OT 1786	10	30
^{117}Sn	OT 1788	235	80
^{118}Sn	OT 1789	175	95
^{119}Sn	OT 1790	220	85
^{122}Sn	OT 1792	120	90
^{124}Sn	OT 1793	170	95
^{69}Ga	PB 182601	100	>99
^{71}Ga	PB 182701	70	98
^{58}Fe	OQ 1779	12	73
^{155}Gd	OZ 182401	3.0	99.8
^{157}Gd	PA 182501	2.6	99.7

ISOTOPIC SEPARATIONS

Stable

The stable isotopes of Cd, Gd, Ga, Fe, and Hg are currently separated on a 5-day around-the-clock basis in 21 beta-type calutrons and three 255°-modified calutrons. The gallium isotope separation replaced a tin separation in a segment of eight calutrons on Nov. 2, 1970, and was completed Jan. 15, 1971. A strontium-isotope separation replaced gallium, and this separation is expected to continue for approximately 9 months in order to obtain about 20 g of ⩾80% ^{84}Sr for isotope sales. The cadmium and gadolinium isotopic separations are expected to be completed around the first of April 1971, while iron and mercury separations are scheduled to continue until current requirements are completed. A hafnium separation will probably follow the cadmium separation and be made in the 255° separators. The use of these machines permits the collection of all hafnium isotopes simultaneously, which was formerly possible only in the alpha machines.

Second-pass separations of ^{155}Gd and ^{157}Gd were completed during October 1970 in the two 24-in.-radius calutrons to provide approximately 3 g of each isotope at greater than 99% purity.

Radioactive

A ^{235}U enrichment program was initiated in four of the contained alpha separators on Oct. 12, 1970, to permit the collection of 500 to 600 g of ^{235}U enriched to >99%. This separation was interrupted, however, on Dec. 1, 1970, after collection of 200 g of ~99.9% material, to separate approximately 1 kg of ^{238}U containing <30 ppM ^{235}U. The ^{235}U separation will be resumed as soon as the ^{238}U objectives have been completed.

NEW GAMMA STERILIZATION PLANTS

International Nutronics, Inc., is operating a gamma sterilization plant at Los Altos, Calif., the first on the West Coast. Products to be sterilized include surgical instruments and cosmetics.

New plants in Europe include Gammaster afd. van de Cooperatieve Apothekers Vereeniging, which is installing a 125-kCi ^{60}Co irradiator (built by AECL), in Utrecht, Netherlands, for sterilizing medical disposables. The plant has a throughput of 4 thousand m^3/year. In England a 1-MCi package-irradiation plant at Reading has been started up by Gamma Sterilisation

XUM

Services Ltd. The ^{60}Co for the $250 thousand plant was supplied by United Kingdom Atomic Energy Authority (UKAEA). In addition, L.R.C. International Ltd. has leased from the UKAEA two gamma-irradiation plants at Wantage, with a total of 700 kCi of ^{60}Co. L.R.C. has formed a new company, Irradiated Products Ltd., to provide a confidential irradiation service to industry, with particular emphasis on sterilization of medical supplies. L.R.C. is also developing and installing a new continuous-gamma-irradiation plant to provide additional facilities for industrial users. The plant is expected to be operational in 1971.

CALIFORNIUM-252

Eighteen universities have been approved by the AEC to receive, on loan, small research quantities of ^{252}Cf. The ^{252}Cf sources, about 1 μg each, are being loaned for use in educational programs—particularly classroom and laboratory demonstrations. Since announcement of the loan program, more than 80 universities have requested application information. The sources to be loaned were fabricated at the AEC Savannah River Plant at Aiken, S. C. They have been used by medical investigators throughout the country and returned to Savannah River upon completion of work under the AEC market-evaluation program for ^{252}Cf.

On Feb. 1, 1971, ^{252}Cf became available at $10 per microgram. The limited research quantities that have been produced in the past have been sold for $100 per 0.1 μg. A standard shipment is 1 mg or more, with a handling charge of $1600 per standard shipment with orders for less than 1 mg and handling charges for them considered on an ad hoc basis. Several companies have notified the AEC of their interest in providing encapsulation services for californium. A list of companies is available from the AEC Savannah River Operations Office, and other companies who intend to provide this service are urged to inform that office.

Additional information on the procedures for purchasing californium, including material specification, a description of the shipping package, and a sales order form, is contained in the "Californium-252 Buyers' Guide." All sales orders, requests for the buyers' guide, and requests for a list of potential californium encapsulators should be sent to the Manager, Savannah River Operations Office, P. O. Box A, Aiken, S. C. 29801.

ISOTOPE PRICES DECREASED

Mound Laboratory has cut the price of 90% enriched ^{13}C to $60 per gram, about 10% of the former price. The Mound prices are $1.75 per gram for 99.95% enriched ^{12}C and $11 per liter for 99.98% enriched ^{16}O.

CONNECTICUT YANKEE TO PRODUCE ^{238}Pu

The ^{238}Pu produced in the AEC Savannah River production reactors may be insufficient for long-term future needs for isotopic energy sources. Southern Nuclear Engineering, Inc., under contract to the AEC, will run tests in the Connecticut Yankee reactor to see if ^{238}Pu with acceptable ^{236}Pu content can be produced by irradiating power-reactor neptunium in light-water reactors. Savannah River is to furnish the 600 g of neptunium oxide needed for the test. Start of the 1-year irradiation was planned for spring 1971.

General

RADIOACTIVITY CALIBRATION STANDARDS AVAILABLE

The proceedings of a special session of the international conference of the American Nuclear Society Meeting on the Constructive Uses of Atomic Energy have been published by the U. S. National Bureau of Standards. Experimental procedures and methods used in nine international radioactivity-standardization laboratories are described. Among the topics discussed are: the possibility of attaining accuracies of the order of 0.01%; the desirability of making international consistency checks for new gamma-radiation solution standards of the same radionuclide issued over a period of time, using a 4π gamma ionization chamber; the validity of radioactivity standards; and an analog method of liquid scintillation counting.

The booklet, *Radioactivity Calibration Standards*, National Bureau of Standards Special Publication 331 ($1.25, SD Catalog No. C13.10:331), edited by W. B.

Mann and S. B. Garfinkel, is available from the Superintendent of Documents, U. S. Government Printing Office, Washington, D. C. 20402.

^{147}Pm-POWERED WRISTWATCH

Seikosha Co. Ltd. of Japan displayed a wristwatch powered by 10 Ci of $^{147}Pm_2O_3$ at the 1970 International Exposition in Osaka. The nuclear battery (Betacel) was obtained from McDonnell Douglas Astronautics Co., U.S.A.

IRRADIATED-FOOD WHOLESOMENESS STUDY

Organizations in 19 countries have agreed to participate in a new international project on the wholesomeness of foodstuffs preserved by radiation. The project, established in January 1971, is sponsored by the International Atomic Energy Agency, the European Nuclear Energy Agency of the Organization for Economic Cooperation and Development, and the Food and Agriculture Organization of the United Nations. The project will be mainly concerned with wholesomeness testing of selected food products treated by irradiation, the tests to be carried out under contract by specialist organizations in member countries of the sponsoring organizations as part of an overall program coordinated by a small central body under the direction of a full-time project leader at Karlsruhe, West Germany.

FIRESTONE TAKES CONTROL OF RADIATION DYNAMICS

The Firestone Tire & Rubber Company of Akron, Ohio, has acquired 65% ownership of Radiation Dynamics, Inc., Westbury, N. Y., an accelerator manufacturer. According to Raymond C. Firestone, Chairman of Firestone, "This acquisition reflects our belief in the potential of the radiation industry, a new field which can lead to manufacturing methods and processes of great advantage to our own company and to industry generally." Among potential industrial applications of radiation are instantaneous curing of plastics, rubber compounds, surface coatings, and laminations; strengthening of materials by providing new bonds between molecules; altering surface or internal properties of semiconductor materials; sterilization of medical goods; rapid change of structure and composition of organic chemicals during processing; and treatment of solid wastes.

NUCLEAR PACEMAKER NOT BLAMED FOR MAN'S DEATH

Although the first man (56 years old) to receive a nuclear-pacemaker implant has died, his death is not attributed to faulty operation of the unit. He had used six conventional heart pacemakers before implant of the radioisotope-powered unit on July 22, 1970, and in October the nuclear pacemaker was replaced by a seventh conventional unit. The postmortem report attributed death to natural causes, with acute failure of the left ventricle due to myocardial conducting insufficiency. His aortic valve had earlier been replaced by a plastic one.

HARWELL ISOTOPE BUREAU

On May 1, 1970, the Harwell Isotope Bureau came into existence at the U. K. Atomic Energy Research Establishment (AERE), strengthening and extending a 20-year-old link with industry. From its beginning, Harwell has worked to develop and promote useful industrial applications for the radioisotopes produced in nuclear reactors, and the Isotope Bureau is being created to maintain and extend the close contacts established with industrial organizations over the past 10 years.

The Bureau will act as a central point for handling inquiries on isotope applications and radiation processing, will actively seek to promote industrial interest in radioisotopic instruments and tracer techniques and in the uses of ionizing radiations, and will seek new ways in which Harwell's special skills and equipment can be used to the best advantage of industry and of the nation.

Further information can be obtained from R. M. Longstaff, Manager, Isotope Bureau, AERE, Harwell, Didcot, Berkshire, England.

NEW CANADIAN RADIOISOTOPE LABORATORY

A radioisotope laboratory has been established at the Canada Centre for Inland Waters in Burlington, Ont., where professionals from Chalk River's Division of Biology and Health Physics, the Department of National Health and Welfare, the Fisheries Research Board, the Association of Universities and Colleges of Canada, and private organizations will conduct studies in hydrology, sedimentation, biology, and physical and chemical limnology using radioactive tracers. Meanwhile, pollution inspectors from the Ontario Depart-

XUM

ment of Health have been flushing $^{32}PO_4$ and other tracers down about 20 cottage toilets in the Cameron Lake area near Fenelon Falls to discover whether sewage is getting into groundwater and lakes in Ontario recreation areas. Samples from groundwater and from the lake near the cottage septic tanks are analyzed to determine whether pollutants are moving down to the water table.

ISOTOPIC GENERATORS ABROAD

An isotopic power generator for use in producing optical and acoustic signals for off-shore oil rigs is being developed in Italy. The direct-conversion system will be produced by the CNEN and the isotopic portion of the system by SNAM-Progetti.

The Japan Atomic Energy Research Institute is working with the University of Tokyo to develop an isotopic generator based on ^{90}Sr. Other radionuclides will be considered later, and the program will include generators for a variety of applications, e.g., heart pacemakers and lighthouses.

NEW X-RAY-FLUORESCENCE SPECTROMETERS ANNOUNCED

An Edinburgh firm, Nuclear Enterprises Ltd., is marketing an X-ray-fluorescence spectrometer that is being used regularly at the National Museum of Antiquities of Scotland to determine the composition of antique objects—e.g., Greek statues, Egyptian rings, and Roman coins—and thus to detect forgeries. For example, allegedly ancient bronzes have been shown to contain copper and zinc instead of the normally expected lead and tin. The equipment is also being used by St. Andrews University in a search for metals in the Southern Uplands of Scotland, and another analyzed moon dust brought back by Apollo 11.*

An automatic X-ray–fluorescence–neutron-activation-analysis system, introduced by Atomic Energy of Canada Limited uses ^{125}I as the source of exciting radiation and a silicon detector in the X-ray part of the system, and a ^{124}Sb–Be neutron source with a germanium detector in the activation-analysis part. The pulse-sorting and data-analysis electronics are shared. The system has applications in pollution control, mineral assay, and rapid identification of steel and nonferrous alloys.

*For a similar instrument developed in the United States, see R. Frankel, Detection of Art Forgeries by X-Ray-Fluorescence Spectroscopy, *Isotopes and Radiation Technology*, 8: 65-68 (1970).

MEDICAL PRODUCTS IRRADIATOR FOR MELBOURNE, AUSTRALIA

Tasman Vaccine Laboratory Limited is locating a new medical-products irradiator in Melbourne, Australia. The Company already operates one at Upper Hutt, New Zealand [*Isotopes and Radiation Technology*, 6: 93-99 (1968)]. The ^{60}Co for the Melbourne unit—initial loading of 200 thousand Ci and capacity of 1.5 million Ci—is being supplied by Atomic Energy of Canada Limited. Startup is scheduled for July 1971.

IRRADIATED FOOD RECEIVED ENTHUSIASTICALLY OVERSEAS

In Israel and The Netherlands, consumers are now able to purchase irradiated foods and—in Israel, at least—are showing a preference for the irradiated product. Dutch housewives can purchase fresh mushrooms either irradiated or unirradiated at the same price. They have accepted the irradiated product easily, and the supermarket chain marketing the treated mushrooms states that the women show no marked preference for either treated or untreated mushrooms. Government officials are expected to approve sale of irradiated strawberries and asparagus next.

In Israel, irradiated potatoes and onions are being marketed at a slightly higher price than the unirradiated products. Customers showed a definite preference for the irradiated potatoes—by a margin of 3 to 1. Some customers even changed from shop to shop to obtain the specially labeled potatoes. The preference for irradiated onions was not as clear-cut, but the consumers were evidently quite satisfied with them.

In both countries a well-planned publicity campaign, backed by the government, preceded the actual sale of irradiated food.

MEDEX-71

MEDEX-71, the first international exhibition and information meeting on medical electronics and bioengineering, was held in Basel, Switzerland, from Mar. 9–13, 1971. The program included discussions on radioisotopic diagnostic methods, clinical scintigraphy with short-lived isotopes, radiation sterilization, and

XUM

problems related to the use of artificial organs. The meeting was both technical and industrial in nature. Information on the meeting may be obtained from Secretariat MEDEX-71, Boite postale CH-4000, Basel 21, Switzerland.

AEC–ORNL ANNOUNCE INTERNATIONAL SYMPOSIUM ON RESEARCH MATERIALS FOR NUCLEAR MEASUREMENTS

Scheduled for October 5–8, 1971, at the Riverside Motor Lodge, Gatlinburg, Tennessee, the referenced symposium will be concerned with a number of topics: (1) Preparation of High Purity Isotopes and Alloys; (2) Coating Technology in Preparation of Targets, "Standard" Sources, and Other Nuclear Research Materials; (3) Self-Supported Film and Foil Preparations; (4) Preparation and Handling of Single-Crystal Metals, Alloys, Compounds, and Films of Separated Isotopes; (5) Problems of Metrology and Assay; (6) Film Nucleation and Growth Phenomena; and (7) a Panel Discussion—Problems of the User Versus the Fabricator. The $25 registration fee covers all activities at Gatlinburg and a preprint of the conference papers. Further information may be obtained from E. H. Kobisk, P. O. Box X, Bldg. 3037, Oak Ridge, Tenn. 37830.

RADIOISOTOPE USE INCREASES IN JAPAN

A 1970 report of the Japan Radioisotope Association shows a 10% overall increase in the number of radioisotope users in Japan in FY 1970—a third of these in industry, a third in hospitals, and the rest in educational, research, and miscellaneous organizations. In some areas growth was especially great; e.g., the number of gamma radiography facilities and isotopic gages in industrial firms increased 23%. Japan is especially prominent in the worldwide medical picture, where she leads the field in ^{60}Co and ^{137}Cs teletherapy units in operation, 3.3 per million population compared to 2.9 for the United States, 2.6 for Canada, and 2.3 for France.

Abstracts of Recent Literature

Manual on Mutation Breeding, Technical Reports Series, No. 119, IAEA, Vienna, 1970 (STI/DOC/10/119) (237 pp; $6.00).

Supplementary Keywords: *mutation; comparison; textbook; agriculture.*

This manual, a joint undertaking of the Food and Agriculture Organization of the United Nations and the International Atomic Energy Agency (IAEA), was discussed as early as 1963, but compilation and editing were not completed until 1970. It is intended for use by advanced university students as well as by plant breeders. Thirty-two experts from 15 countries contributed 40 chapters on such subjects as the use of mutations in plant-breeding programs, radiation and chemicals as mutagens, causes and nature of mutations, problems of mutation breeding, mutagen effects, specific characters to be improved by mutation breeding, mutation breeding of vegetatively propagated plants, and when to use mutations in plant breeding. The bibliography contains about a thousand items. The manual is being supplemented by reviews on specific crops, by Å. Gustafsson of Lund, Sweden; the first of the series, "Mutations and Crop Improvement, I to VII," was published in *Hereditas,* 1965–1966. (MG)

Nuclear Techniques for Increased Food Production, Food and Agriculture Organization, United Nations, Basic Study No. 22, Rome, 1969; available from UNIPUB, Inc., P. O. Box 433, New York 10016.

Supplementary Keywords: *agriculture; food preservation; tracer; mutation; insect control; hydrology; review, technical.*

The many ways in which radiation and radioisotopes have contributed to increase the world's food supply are reviewed in this joint FAO/IAEA publication. The material is up-to-date and complete, but there are no references to guide the reader deeper into any of the subjects presented.

Part I shows how radiation has increased crops through induced mutations, such as changing the color of Sonora 64 wheat so that Indian consumers would accept it. The sterile-male technique, with emphasis on

eradication of the fruit fly in Central America, is covered in detail. Animal vaccines produced by radiation attenuation of the parasite in question are now available. The coverage of the use of radiation to preserve foods includes a table, with an addendum on the last page of the report, which summarizes worldwide approvals by health authorities for marketing irradiated food.

Part II relates how radioisotope tracers have been used to evaluate fertilizers and study plant–soil relations. They have also been used to study how animals adapt to new environments. Radioisotope tracers help protect the available food supply by enabling scientists to detect toxic chemicals and to determine how pesticide residues in food behave.

Part III deals with the important subject of water. Tracers have been used to study underground water movement; radiation sources can be used to determine the moisture content of soil. There is also a section on nuclear desalting.

Nuclear techniques bear upon all phases of agricultural practice. They are not substitutes for existing techniques but are valuable additions to the arsenal of methods being applied to prevent the death and disease caused by insufficient food supplies. (FEM)

Isotopes and Radiation in Parasitology II, Proceedings of Research Coordination Meeting, Vienna, June 2–6, 1969, IAEA, Vienna, 1970 (STI/PUB/242) (139 pp., $4.00). See also meeting of July 31–Aug. 4, 1967, IAEA, Vienna, 1968 (STI/PUB/181) (157 pp., $3.50), summarized in *Isotopes and Radiation Technology,* **6**: 306-317 (1969).

Supplementary Keywords: *conference; zoology; veterinary; source, gamma; accelerator; tracer; modification, material;* ^{3}H; ^{51}Cr; ^{60}Co; ^{125}I.

A meeting was held in Vienna in 1969 to review progress, since the last conference, in studies of parasitic diseases in domestic animals. The proceedings contain 17 papers and the recommendations of the research coordinators.

Urquhart and Jarrett (Scotland) found that rats did not develop the typical immune reaction when infested with worms unless the rats were over 6 weeks old. They conclude that vaccination will be needed during this immunological immaturity period and that initial doses of irradiated larvae should be larger. Miller (Scotland) and coworkers (United States) tested an irradiated canine-hookworm vaccine; the inoculated pups were unaffected by a challenge that severely affected controls and depressed their hematologic values. A similar experiment with guinea pigs was run by Tomanek (Czechoslovakia). Although the use of irradiated larvae did confer immunity, subcutaneous inoculation with unirradiated larvae gave greater protection. Tomanek and Prochazka (Czechoslovakia) tested the effectiveness of an X-ray machine and an accelerator. Twice the radiation dose was required from the accelerator as from the X-ray machine to obtain the same degree of larval attenuation. The behavior of irradiated and unirradiated metacercariae of *Fasciola hepatica* in rabbits was studied by Movsesijan and Cuperlovic (Yugoslavia). They found that radiation attenuated the pathogenic effect but preserved the antigenic effect at doses of 2 kR; larger doses inhibited development of the parasite and did not lead to development of antibodies.

Bitakaramire (Kenya) attempted studies on cattle infected with *Fasciola gigantica,* using ^{51}Cr-labeled red cells and ^{125}I-labeled albumin, but the experiment did not lead to clear differences between infected and control animals. Other studies of fluke infections were reported by Dargie and associates (Scotland). The onset of pathophysiological effects coincided with establishment of parasites in the bile duct. Results confirm that the primary cause of anemia and hypoalbuminemia is blood loss into the gut via the bile.

A field trial of irradiated gapeworm larvae showed that only 2.8% of 527 protected chickens died, as compared with 74.5% of 342 controls, reported Varga and Vetesi (Hungary). The hypoproteinemias associated with helminthic diseases were studied with isotope techniques by Nielsen and Nansen (Denmark). Plasma half-lives are related to the size of hepatic fluke infection, which indicates a biliary loss of albumin. The synthesis rate of immunoglobulin is increased, however, even though this immunological response may not actually show that immunity is developed. Nansen (Denmark) also reported that use of radioiodinated immunoglobulin in cattle and sheep revealed a marked increase in synthesis, again without affecting the flukes. Hurwitz (Israel) presented a model for study of intestinal digestion and absorption in the presence of helminthic infections, using a nonabsorbed reference substance labeled with ^{91}Y.

Duxbury and Sadun (United States) induced protection against African trypanosomiasis with inoculations of radiation-attenuated trypanosomes in mice and rats. Doses of 20 kR were sufficient to protect the host animals. The immunity in rats did not last, however. Irradiation of *Plasmodium*-parasitized erythrocytes with doses of 20 krads led to a resistance in rats inoculated with the product, according to Sadun and

XUM

coworkers (United States). An important cattle disease in Africa is East Coast fever, caused by the parasite *Theileria parva.* A technique for isolating infective particles from the intermediate tick host was given by Purnell and associates (Kenya). Autoradiography with ^{3}H was used by Soulsby and Muncey (United States) to study lymphoid cell reactions to tissue nematodes. Cell-mediated immunity is the primary response in the liver; humoral antibody production occurs in the lungs.

Casarosa (Italy) proved that animals vaccinated against *Ascaris* but given an $LD_{50/30}$ dose of radiation died more rapidly than nonirradiated, vaccinated animals given a lethal dose of the parasite. Sokolic and associates (India) indicate that *Dictyocaulus filaria* infection of sheep and goats, which is serious in India, could be controlled by application of the radiation vaccine developed in Yugoslavia.

The panel recommended that the IAEA promote the use of radiation-attenuated vaccines, specifically by informing member states of their availability. Further studies are needed on better production methods and incidence of the diseases. The mechanisms of immunity to parasitic diseases should be studied in detail; methods for using radioisotope tracers were proposed. Four specific protozoal diseases were judged sufficiently prevalent to warrant much intensive research: trypanosomiasis, malaria, coccidiosis, and East Coast fever. Finally, more work is needed for elucidation of the pathophysiology of parasitic infections. (FEM)

Power and Research Reactors in Member States—May 1970 Edition, IAEA, Vienna, 1970 (STI/PUB/194/3) (60 pp., 1 removable map; $2.00).

Supplementary Keywords: *industry, nuclear energy; data compilation.*

The IAEA has published the third issue of its biannual comprehensive list of land-based nuclear-power and research reactors. The list is preceded by definitions of abbreviations pertinent to reactors and by numerical tables and corresponding graphs that show, by country, the projected growth in installed capacities of power reactors (until 1976) and in the number of research reactors (to February 1970). For power reactors, the list is arranged in three groups according to power output. For each group—and also for research reactors—those reactors in operation and those under construction are listed separately. A reactor name index in two parts (Power Reactors and Research Reactors) is given. A wall-size removable map of nuclear power stations in member states of the IAEA is a bonus. (HPR)

Isotope Techniques for Studying Animal Protein Production from Nonprotein Nitrogen, Report of Consultants' Group Meeting, Vienna, Aug. 25–28, 1969, (FAO/IAEA), IAEA, Vienna, 1970 (STI/DOC/10/111) (29 pp., 46 references; $1.00).

Supplementary Keywords: *biochemistry; industry, food; material tracing; flow measurement; comparison; review, technical; bibliography;* ^{3}H; ^{14}C; ^{15}N; ^{24}Na; ^{32}P; ^{35}S; ^{40}K; ^{42}K; ^{51}Cr; ^{144}Ce.

For some countries, two-thirds of the population's daily protein intake is from animal origin. As people become more prosperous, protein intake from this source will increase. Among food-producing animals, ruminants are unique in being able to convert poor-quality protein and nonprotein nitrogen (NPN) into high-quality protein for human food. The prospect exists that cereals and other high-carbohydrate feeds may become available to support ruminant production of meat and milk. Thus, the possibility of supplementing these protein-deficient feedstuffs with nonprotein nitrogen should be investigated.

The role of isotopes in this investigation was the subject considered by a Consultants' Group of 25 scientists from 13 countries. This IAEA publication sets forth the results of their discussion of the evaluation of NPN sources in feed, the direct evaluation by animal-retention studies, indirect assessment and predictive methods (protein synthesis in and beyond the rumen), the use of ^{15}N for the study of NPN, and analytical techniques. Their conclusions regarding the possible application of isotopes to research on evaluating NPN in ruminate rations follow:

Isotope	Application
^{15}N	Rate of NH_3 production in rumen
	Rate of incorporation of N compounds into microbial protein
	Overall conversion of NPN to tissue or milk protein
^{14}C	Rate of hydrolysis of C-containing NPN
	Turnover and entry rate of amino acids
	Estimates of protein synthesis
^{35}S	Estimates of microbial protein synthesis
	Estimates of microbial contribution to synthesis of milk, wool, muscle
^{2}H, ^{3}H, ^{40}K, ^{13}N	Body composition
^{51}CrEDTA and ^{144}Ce	Indicator to study digesta flow rates, dilution rate, and extent of absorption of amino acids

Three of the five recommendations to FAO and IAEA by the Consultants' Group involve isotopes; they are that:

- The Joint FAO/IAEA Division of Atomic Energy in Food and Agriculture coordinate worldwide studies that involve the use of isotopes in NPN research designed to increase animal-protein production.
- Research programs involving the use of isotopes be coordinated further to study all phases of animal-protein synthesis and metabolism in domestic animals.
- The Joint FAO/IAEA Division of Atomic Energy in Food and Agriculture make available information on the use of isotopes for NPN research with particular emphasis on ^{15}N. Further, it is suggested that facilities be made available—preferably at the IAEA laboratory— for training in ^{15}N techniques, especially those involving the relatively new method of emission spectroscopy. (HPR)

Neutron Moisture Gauges, A Guidebook on Theory and Practice, Technical Reports Series, No. 112, IAEA, Vienna, 1970 (STI/DOC/10/112) (95 pp., $3.00).

Supplementary Keywords: *measurement, moisture; source, neutron; textbook.*

The participants in a 1966 Vienna meeting on neutron moisture gages recommended preparation of a manual on the theoretical and practical aspects of these gages, which have been in use since 1950 or earlier. A panel of experts from six countries compiled, edited, and ensured updating of the material. Chapters are included on the characteristics, physical bases, design, accuracy, calibration, and selection of neutron moisture gages, as well as on health and safety characteristics. Appended are proposed definitions of 32 pertinent terms, which, hopefully, will form a basis for international usage. All chapters are documented, with a total of about 100 references. (MG)

Sterilization and Preservation of Biological Tissues by Ionizing Radiation, Report of a panel meeting in Budapest, June 16–20, 1969, IAEA, Vienna, 1970 (STI/PUB/247) (129 pp.; $4.00).

Supplementary Keywords: *prosthetic device; accelerator; ^{60}Co.*

Current practices in transplantation of organs and tissues have led to investigation of new methods of sterilization. Twenty-one radiobiologists, surgeons, and specialists in irradiation techniques from nine countries met in 1969 to discuss the use of large radiation sources for sterilization of these materials. Both radioisotopic and machine irradiation were included. The basic principles of the technique were considered, along with the effects of radiation on tissues. Specific tissues whose preservation was discussed included bone, cartilage, heart valves, and the aorta and other blood vessels. The 11 papers presented are individually documented, with a total of about 160 references. (MG)

Isotope Hydrology 1970 (Proceedings of a Symposium, Vienna, Mar. 9–13 1970, organized by IAEA in cooperation with UNESCO), IAEA, Vienna, 1970 (STI/PUB/255) (917 pp., 54 papers; $24.00).

Supplementary Keywords: *ecology; sedimentology; logging; material tracing; activation analysis; leak detection; flow measurement; isotope abundance; absorptiometry; instrument; portable; bibliography.*

International and interdisciplinary cooperation is totally essential to the development of water resources—one of the most urgent worldwide problems. Isotope hydrology is fast becoming a most useful tool in this work.

The third symposium on the use of isotopes in hydrology reflects the growing interest in the subject through the large increase in the number of papers and amount of discussion. The participation of both practicing hydrologists and isotope specialists from 44 countries indicated that the earlier research has developed into practical working methods of worldwide interest. The session topics and number of papers presented on each were

Environmental Isotopes in the Water Cycle	13
Problems on Carbon-14 Dating of Groundwater	5
Field Studies Using Environmental Isotope Techniques	8
Tracer Techniques in Surface Water Studies	7
Sediment Studies	3
Borehole Techniques and Nuclear Instruments	6
Leakage from Reservoirs and Canals	4
Computational Studies	3
New Techniques in Tracing and Dating	5

These topics are much more varied than those of the second symposium, which were included in six general areas. Those of the first symposium were still more limited—primarily to flow measurement, especially of groundwater. Thus, the ever-broadening scope of the use of isotopes in hydrology is evident.

Isotope Hydrology 1970 makes available the new information on theoretical and experimental developments that led to the solution of practical problems in

XUM

hydrology. The publication should achieve its intended goal—to speed up the adoption of these new isotopic techniques. (HPR)

Nondestructive Methods of Test for Concrete: Part 3, Gamma Radiography of Concrete (BS 4408), British Standards Institutional Sales Branch, 101/103 Pentonville Road, London N.1 (12 shillings).

This publication of the British Standards Institution presents a method for radiography of hardened concrete which can detect such faults as incomplete compaction, cavities in the grouting of prestressing wires, and defects in joints between concrete units, and can accurately determine the position of internal steel reinforcement. The techniques are based on current good practice using procedures that have been used successfully in laboratory and field studies. Concrete as thick as 45 cm can be examined. (MG)

Microbiological Specifications and Testing Methods for Irradiated Food, Panel Report, IAEA, Vienna, 1970 (STI/DOC/10/104) (supplement to *The Technical Basis for Legislation on Irradiated Food,* Joint FAO/IAEA/WHO Committee, Rome, April 21–28, 1964, FAO, United Nations Atomic Energy Series Report No. 6; also available as World Health Organization Technical Series Report No. 316, 1966).

Supplementary Keywords: *microbiology; conference; food preservation; industry, food; standard.*

An expert panel has prepared a supplement to an earlier FAO publication on the technical basis for legislation on irradiated food. Microbiological methods for irradiated food were excluded from the earlier publication. A panel met in Vienna in June 1965 to discuss methods and to assemble information on formulations, ingredients, and techniques. A significant work on microbiological methods was subsequently completed by the International Committee on Microbiological Specifications for Foods (ICMSF) of the International Association of Microbiological Societies. At a second meeting of the expert panel, this work was incorporated into a final report, and the number of methods proposed for the evaluation of food for specific microorganisms was reduced.

Clarity is essential in formulating legislation or regulations, particularly so in microbiology where the values obtained for the microbial content of a food are likely to vary widely. If a regulation specifies that certain microorganisms are permitted only in certain quantities, the methods and techniques used to comply with the regulation must also be specified. The panel experts compared the methods being used at present, reviewed the scientific literature, and pooled their experience and knowledge to consolidate worldwide microbiological information into a limited number of acceptable, reproducible methods that could form the basis for evaluating the effectiveness of different levels of radiation in processing food and feeds, the microbiological quality of the resulting products, the microbiological safety of the products, and the closeness with which regulations should comply with microbiological requirements.

The report is divided into four parts: (1) microbiological specifications, (2) recommended methods for microbiological analysis of irradiated foods, (3) culture media, and (4) reagents. In addition, a list of recommendations for further research is given. In Part 1, some problems of special importance in irradiated foods are discussed and general specifications for irradiated foods are given. The quality of foods to be irradiated must be high since irradiation is not a substitute for proper hygienic food handling. Packaging requirements are also set out in this part. Labeling requirements are established to attain three objectives: the consumer must be able to exercise a preference for either irradiated or nonirradiated food; data must be provided to authorities investigating public-health problems; and import authorities must be able to determine that a product has, in fact, been irradiated. Specifically, labels must show the actual date of irradiation, the type of irradiation treatment (including dose range), and a recommended storage temperature. The panel felt that specific limits of shelf life for specific foods could not be established because information on which to base such recommendations is not yet available. However, they did feel justified in expressing the opinion that microbiologically unstable foods, such as perishable meat and poultry products, cooked meats, bakery products, and other ready-to-eat foods should not be treated by radicidation. Specifications for particular types of radiation treatment to different categories of food are set out; these include requirements for processing data, microorganisms of special interest, packaging, requirements for testing, and limitations of each process.

In Part 2, the recommended methods for microbiological analysis of irradiated foods are presented in detail. The adoption of standardized methods has been a long-recognized need and could well prove to be a major aid to international acceptance of irradiated foods. The panel accepted a selection of certain methods recommended by the ICMSF and endorsed

XUM

their statement that these methods were to be considered interim methods, with improvement and simplification expected after appropriate laboratory testing. The details of the methods comprise nearly half the report.

Parts 3 and 4 are compilations of proper laboratory procedures used to prepare and maintain the culture media and reagents necessary for the tests detailed in Part 2. A list of 70 references is given, and the report concludes with specific recommendations by the panel of further research needed on the microbiology of irradiated foods. (FEM)

Tracer Diffusion Data for Metals, Alloys, and Simple Oxides, by John Askill (Millikin University), IFI/Plenum Data Corporation, New York, 1970 (107 pp., 473 references; $12.50).

Supplementary Keywords: *metallurgy; chemistry, physical; material tracing; physical property; bibliography; textbook; review, technical; data compilation; theory, applied.*

Radioactive-tracer-diffusion data for metals, which have been reported over a 30-year period (1938–1968), are compiled in *Tracer Diffusion Data for Metals, Alloys, and Simple Oxides.* Although atomic diffusion in metals was discovered some 65 years ago, work done before about 1940 is chiefly of historical interest because of uncertainties in the measured diffusion coefficients. The availability of high-specific-activity radioisotopes after World War II resulted in a tremendous advance in diffusion studies. It became possible to measure self-diffusion rates and to study diffusion of impurities present in concentrations too small to change the chemical homogeneity of the system.

Diffusion-rate data are important in processes controlled by rates of atomic migration, such as growth of phases and homogenization of alloys, and also in theories of such phenomena as oxidation, plastic deformation, sintering, and creep. In an introductory chapter, the various aspects of the diffusion data, given later in the tables, are discussed; these aspects include temperature range, method of analysis, and the diffusion parameters D_0 and Q of the Arrhenius equation $D = D_0 \exp(-Q/RT)$. The mechanisms proposed for atomic diffusion in solids are described. In a second introductory chapter, the empirical relations for diffusion, especially self-diffusion, are discussed. From these relations, it is possible to estimate approximately the diffusion coefficient at some temperature in most systems for which direct experimental data are not available.

The data compilations (50 pages), which follow the introductory chapters (29 pages), are in four parts:

I. Self-diffusion in pure metals.
II. Impurity diffusion in pure metals.
III. Self- and impurity diffusion in alloys.
IV. Self- and impurity diffusion in simple metal oxides.

The compilations include more than 1200 entries; elements and solutes are arranged alphabetically, and the callout number and year of the reference are given with each entry. The radioactive tracers used are listed in the first column. Author and brief subject indexes are included.

The book is comprehensive (473 references) yet concise (107 pages). Dr. Askill has contributed a reference volume that should be exceedingly time-saving and valuable to those concerned with diffusion in solids whether their interest be in data already reported or in studies they are planning. (HPR)

Training Manual on Food Irradiation Technology and Techniques, IAEA, Vienna, 1970 (STI/DOC/10/114) (134 pp., 45 references; $2.00).

Supplementary Keywords: *education; industry, food; food preservation; textbook; bibliography.*

In over 50 countries, research is being done on food preservation by ionizing radiation—the only novel approach to preserving food since canning was invented 150 years ago. To provide needed technologists, the IAEA, FAO of the United Nations, and the USAEC jointly offered training courses in 1967 and 1969. The success of the courses prompted the IAEA and the FAO to prepare this manual, which consists of basic information and laboratory exercises.

The basic part (Part I, Lecture Matter) is covered in 13 chapters: (1) Radioisotopes and Radiation, (2) Radiation Detection and Measurement, (3) Health Physics, (4) Radiation Chemistry, (5) Effects of Radiation on Living Organisms, (6) Preservation of Foods, (7) Radiation Preservation of Foods, (8) Packaging, (9) Wholesomeness of Irradiated Foods, (10) Government Regulation of Irradiated Foods, (11) Food Irradiation Facilities, (12) Commercial Considerations of Food Irradiation, and (13) Literature Sources. Each chapter contains carefully selected material that is specifically useful in food-irradiation technology.

The other part (Part II, Laboratory Exercises) contains nine well-designed experiments that demonstrate how food is irradiated and what effects the irradiation produces in the food product. Exercises are

XUM

provided in the following aspects of food irradiation: (1) dosimetry, (2) radiation chemistry, (3) radiation microbiology, (4) preservation of foods, (5) sensory evaluation of foods, and (6) control of insect infestation. Many of the experiments can be done independently of an instructor.

The manual should result in a better international understanding of irradiation preservation of foods. It should be extremely useful not only to IAEA and FAO trainees but also to any scientist in any country concerned with developing food-preservation technology or introducing irradiation processes into a food industry. (HPR)

Determination of Steroid Hormones, by Ian F. Sommerville, Medical Monograph No. 7, The Radiochemical Centre, Amersham, England (48 pp.).

The literature (168 references) on quantitative determination of steroids using labeled compounds is discussed with regard to use of ^{35}S-, ^{125}I-, ^{131}I-, ^{3}H-, and ^{14}C-labeled reagents for double-isotope-derivative formations; of ^{3}H- and ^{14}C-labeled steroids as internal standards and in competitive protein-binding techniques and radioimmunoassay; and of labeled steroids in studying steroid metabolism. (MG)

Resistance Tests for Sealed ^{60}Co Teletherapy Sources (in French), report of Laboratoire National d'Essais to CEN at Saclay.

At the request of the CEN at Saclay, the French National Testing Laboratory made tests on the CEA sealed ^{60}Co teletherapy sources to define conditions of use under which these sources maintain their integrity. The tests included exposure to both heat and mechanical shock—impact, vibration, perforation, etc. The report gives a detailed description of the sources (from the CEA 1968 catalog) and of the tests and records the results, which show that the stainless-steel source capsules used are satisfactory. (MG)

Application of Radiation Polymerization for the Production of Water-Insoluble Enzyme Preparations, by J. Dobo, Plastics Research Institute, Budapest, *Acta Chimica Academiae Scientiarum Hungaricae,* **63**: 453-456 (1970).

Preparations in which an enzyme is bound to an insoluble carrier, for use as heterogeneous specific catalysts, have been made by an irradiation technique. The monomer acrylamide was X irradiated (X-ray machine) in the presence of trypsin, as a result of which the monomer polymerized to a water-insoluble gel, entrapping the enzyme. With radiation polymerization, heating of the preparation can be controlled more easily than when the reaction is induced with heat. Also, no extraneous foreign material remains in the preparation, as usually occurs when chemical agents are used. (MG)

Tracer Studies in Fresh-Water Streams, by J. C. Sorensen and C. B. Smith, USAEC Report UCLA-ENG-7088, University of California, Los Angeles, September 1970.

Isotopic tracers—both radioactive and stable—were used in studying stream dynamics in an effort to trace the transport and dilution of pollutants in the stream water. The radioactive isotope used was ^{24}Na as sodium carbonate. Three milliliters of a dilute tracer solution was introduced into the stream, and the ^{24}Na appearing at several points downstream was counted at intervals by a submerged gamma counter. The stable isotope used was ^{127}I as potassium iodide. Water samples were taken downstream, and the amount of iodine present was determined by activation analysis. The data indicated no holdup of tracer in the part of the stream studied, and tagging of sources of pollution with isotopic tracers is recommended for future stream studies. (MG)

Wood–Plastic Composite Used in French Parquet Floor Restoration (in French), *CEA Notes d'Information,* No. 7: 5-7 (July 1970).

Supplementary Keywords: *polymerization; artifact; industry, building; industry, wood; source, gamma; ^{60}Co.*

The Radioelement Application Section of the French Atomic Energy Commission has cooperated in the use of an irradiation technique to make a wood–plastic composite in the restoration and preservation of an 18th Century parquet floor in a Grenoble museum. The 30-mm-thick hardwood floor—oak, cherry, sycamore, and walnut—was attached to a resinous backing and had been badly damaged by humidity, insects, and, more recently, women's sharp heels.

The floor area was 155 m^2, with a number of 45- by 45-cm squares bordered along the four walls with strips, each 3 m long and 0.5 m wide. Preliminary studies were made of the woods to determine whether or not the proposed treatment would affect the dimensions.

The treatment consisted in placing the original squares, loaded in four carriers, in an impregnation tank, which was held at a partial vacuum (10 to

XUM

20 mm Hg) for 4 to 5 hr. The monomer—methyl methacrylate, selected because of ease and speed of treatment with it and the short time in which the work had to be done—was then introduced, and the tank was held under 5 to 6 atm nitrogen pressure for 16 hr. Excess monomer was drained off, the tank was opened, and the pieces, in polyethylene sacks under a nitrogen atmosphere, were exposed to the gamma radiation from a 100-kCi ^{60}Co source. The large panels were irradiated flat, with pressing, to prevent deformation during polymerization.

One month was required to process 4500 kg of wood–plastic composite. There were no difficulties during reinstallation, and the carpenters quickly became accustomed to the special properties of this new material. (MG)

Application of Radioisotopes with Magnetic Flowmeters for the Determination of the Solids Content in Pulps in Closed Pipe Systems (in German), *Aufbereitungs-Technik*, No. 9: 549-555 (1965).

With land becoming increasingly unavailable and with standards for water purity being increased, proper management of a settling pond is essential to any ore-processing or limestone-handling plant. The Dornap Cement Works, West Germany, found that the quantity of solids in the slurry leaving the preparation plant was not what the raw-materials-preparation department had predicted; the buildup rate in the settling pond deviated considerably from predicted values. The problem of measurement of solids content was solved by application of an unusual combination of instruments—a 100-mCi ^{137}Cs density gage and magnetic flowmeters. Measurements are now made with an accuracy of 2 to 3%. Closer control of solids content will result in better planning for settling-pond use and enlargement. (FEM)

New Use of Short-Lived Radionuclides—Chemical Rate Measurement, *Chemical Engineering News*, p. 33, Sept. 28, 1970.

Supplementary Keywords: *tracer; reaction mechanism; comparison; conference;* ^{217}*At.*

The method of chemical-nuclear-rate coupling uses nuclear decay rates to measure chemical rates by incorporating a short-lived radionuclide in the reaction. J. Robb Grover of Brookhaven National Laboratory is making a prototype experiment to demonstrate the method. A molecular beam of HAt made with ^{217}At (0.032-sec half-life) is crossed with a beam of Br atoms; the reaction product At and scattered HAt are detected with chemically selective detectors. The method differs from radioactive tracer techniques (provides efficient detection), from hot-atom chemistry (exploits energetic species that result from the dynamics of nuclear transitions), and from many detection methods. The main advantage of the chemical-nuclear-rate coupling method is its high signal-to-noise ratio. The ratio is high because it is independent of time—nuclei birthrate equals decay rate—and because the rapid nuclear decay strongly suppresses noise. The more than 130 radionuclides available from 69 elements, with half-lives from 10^{-5} to 10^{-1} sec, give the method potentially wide applicability. (HPR)

Absolute Stereochemistry of the Second Step of Ribonuclease Action, *Nature*, 228: 663-665 (1970).

Decomposition of a ribonucleic acid is catalyzed by pancreatic ribonuclease in a two-step process—transesterification followed by hydrolysis of the resulting cyclic phosphate. The suggested mechanisms are either "in-line" or "adjacent" depending on the relative geometry of the incoming and outgoing groups during the reaction.

D. A. Usher and D. I. Richardson, Jr., of Cornell University used two diastereoisomers of uridine-2′,3′-cyclic phosphorothioate previously isolated by F. Eckstein of Max-Planck Institute for Experimental Medicine, Goettingen, Germany. Hydrolysis of one diastereoisomer by pancreatic ribonuclease A in ^{18}O-enriched water gave an acyclic ester of fixed configuration, which, on subsequent nonenzymic ring closure, gave the two isomers with ^{18}O only in the isomer for which an in-line ring opening predicted incorporation. (HPR)

^{14}C Used To Compare Metabolism in Man and Animals, *Chemical Engineering News*, pp. 44–45, Sept. 28, 1970.

Supplementary Keywords: *biochemistry; biology; medicine; tracer; material tracing; reaction mechanism; chromatography; conference.*

With ^{14}C, an in-vitro technique was developed for metabolic studies which reflects qualitatively the in-vivo metabolic processes of animals, including man, under prescribed conditions. Thus, the risk of dosing humans may not be required to predict chronic toxicity to man of a compound under study. The technique may eventually lead to the selection of the animal species most similar to man for long-term toxicity studies.

XUM

The pesticide, Carbaryl, was studied by L. J. Sullivan, B. H. Chin, and C. P. Carpenter in work sponsored by Carnegie-Mellon University and Union Carbide Corporation. They used 1-naphthyl methyl-^{14}C-carbamate (Carbaryl), 1-naphthyl-^{14}C methyl-carbamate, and 3,4-dichlorobenzyl-^{14}C methyl-carbamate as parent compounds. The metabolism of organ sections removed aseptically from rats, beagles, and guinea pigs was supported on Trowell T8 medium under medical-grade oxygen in gas-tight culture chambers. A human-liver biopsy specimen was also included. Metabolic profiles of the culture medium, which was shown to have the same distribution of metabolites as the tissue homogenates, were obtained by DEAE cellulose column chromatography. Comparison of the radiochromatogram of the medium with in-vivo results based on 24-hr urine collections showed that the method is qualitatively correct and is also semiquantitative. The in-vitro metabolic-product distribution varies from species to species.

The Carnegie-Mellon group caution that the technique should not replace existing procedures for evaluating toxicity of chemicals. Rather, they consider that a workable system has been devised that can be refined and developed to study the metabolism of many currently confusing physiological responses.

(HPR)

Radioisotope X-Ray Fluorescence Spectrometry, Report of a Panel Held in Vienna, May 13–17, 1968, Technical Reports Series, No. 115, IAEA, Vienna, 1970 (STI/DOC/10/115) (104 pp., $3.00).

The Panel whose findings are reported here reviewed radioisotopic X-ray fluorescence techniques and applications. Their presentations and discussions indicated that reluctance on the part of labor and management to accept a radioactive source as an integral part of a commonplace instrument is a barrier to the rapid introduction of these instruments in industry in some countries. The accuracy and limits of detection of analytical instruments based on this technique are governed by interelement effects in alloys and by interelement, particle-size, and heterogeneity effects in minerals. The lack of suitable standard materials precludes intercomparison of instruments on an international basis and is thought to deter a more rapid acceptance of these instruments, especially in the mining field.

Furthermore, more accurate fundamental data, especially attenuation and excitation coefficients for low-energy X rays, are required to improve the accuracy of performance calculations. Ninety-six references and descriptions of several manufacturers' instruments are appended. (MG)

Radioisotope Production from Fission Products: Review of the Literature, by H. Amano, H. Nakamura, M. Kubota, S. Takashi, and S. Tachimori, Japan Atomic Energy Research Institute, Report JAERI-4051, 1970.

This report (in Japanese) will be of interest to all persons interested in radioisotope production, even those who do not read Japanese, because of the extensive reference list (approximately 300 titles) and the compilation of flow sheets, tables, diagrams, and decay schemes, which are in English. The radionuclides considered are the important short-lived products ^{99m}Tc, ^{132}I, and ^{133}Xe and the parents of the first two, i.e., ^{99}Mo and ^{132}Te. Recent papers on chemical separation methods are reviewed, and information on the behavior of ^{131}I is assembled for use in preventing air pollution. Finally, large-scale production, with emphasis on chemical separation of fission products, is reviewed. (MG)

Chlorophyll Metabolism in Green Plants, by A. A. Shlyk, Izdatel'stvo Nauka i tekhnika, Minsk, 1965, T. V. Godnev (Ed.); translated from the Russian by B. Eisenkraft, USAEC Report AEC-tr-7094 (293 pp., 491 references).

Supplementary Keywords: *agriculture; botany; biochemistry; material tracing; reaction mechanism; review, historical; review, technical; ^{14}C; ^{15}N.*

Chlorophyll Metabolism in Green Plants is a fascinating account of the unraveling of the chemistry of chlorophyll biosynthesis. The author's own experimental work that was done at the Biological Institute and the Laboratory of Biophysics and Isotopes, both of the Academy of Sciences of the Belorussian SSR, is emphasized. In addition, and as far as possible, the directly relevant results of all investigators—whether in agreement with Shlyk's or not—are cited (491 references!).

The purposes of Shlyk's experiments were to elucidate the mechanism of chlorophyll synthesis in green leaves and to find similarities and differences in the situation in etiolated (grown in the dark) sprouts; determining the origin of chlorophyll *b* was of particular interest. The necessity to follow the synthesis of new molecules in the presence of an existing pigment pool required the use of radioactive tracers and thus

XUM

resulted in the first kinetic studies in this field with radioisotopes (^{14}C and ^{15}N).

The book describes the available evidence of chlorophyll metabolism in green leaves, analyzes the biosynthetic relations between the pigments, and relates biosynthesis with the state of the molecules. The first chapter describes the determination of the specific radioactivity of chlorophyll, gives evidence of chlorophyll turnover in the green plant, and analyzes chlorophyll synthesis in detail. Chapter II tells how ^{14}C and ^{15}N were used to reveal the roles of chlorophyll *a* and chlorophyll *b*; chlorophyll *a* precedes chlorophyll *b* in the biosynthesis pathway and can be converted to chlorophyll *b* in the absence of light. These were the first data to support the idea that newly synthesized molecules of chlorophyll *a* are the major precursor of chlorophyll *b*. Chapter III deals with the assembly of components (phorbin and phytol) in the biosynthesis of chlorophylls *a* and *b*. The precursors of phorbin (protochlorophyllide and chlorophyllide) and their metabolic roles as intermediates are discussed in Chaps. IV and V. Chapter VI is devoted to kinetics; the rate of chlorophyll metabolism in green leaves is estimated, and the idea of the heterogeneity of chlorophyll is developed. This idea is confirmed in Chap. VII, where chlorophyll's heterogeneity is shown to have consequence on its metabolism and where the greater lability of chlorophyll *a* relative to chlorophyll *b* is discussed. The book ends with well-presented conclusions regarding seven unknown or disputed features of chlorophyll metabolism.

The book is a highly significant contribution to the field of photosynthesis—a main subject in modern biology. (HPR)

XUM

Index

Isotopes and Radiation Technology

Volume 8

The page range for each of the four issues of Vol. 8 is as follows:

No. 1, pages 1 to 136
No. 2, pages 137 to 252
No. 3, pages 253 to 372
No. 4, pages 373 to 475

A

Accelerators
  electron, design activities in the USSR, 222
Accelerators, Inc.
  sale of minority interest to Hughes Aircraft, 241
Aclar 191 films
  diffusion of H_2O, Na ions, and Cl ions through, radiochemical study of, 60–64
Advisory Committee on Isotopes and Radiation Development
  membership appointments, 368
AEC Critical Review Series, 134, 250, 372
AEC Symposium Series, 135–36, 250–52
Agriculture
  list of improved crop mutants produced by radiation, 96–101
  use of radioisotopes, publication on, 466–67
Aluminum
  determination in Chicago air by neutron activation analysis, 460
American Radiochemical Corporation
  production and sale of labeled compounds, 241
Americium-241
  price reduction, 243
  use in lightning rods for air ionization, 178–80
Amino acids
  determination in proteins by X-ray photoelectron spectroscopy, 369
Animal science
  use of C in metabolism studies, 473–74
  use of radioisotopes in studying protein production from nonprotein nitrogen, report on, 468–69
  use of radioisotopes in, training course on, 365
Antimony
  determination in Chicago air by neutron activation analysis, 460
Antimony-121
  future availability, 240, 364
Antimony-123
  future availability, 240, 364, 462
Apricots
  radiopreservation, 80
Archaeology
  ^{14}C dating of iron in, 320
Argon
  properties of, 255
Art pieces
  forgeries, detection by X-ray-fluorescence spectroscopy, 65–68
Atlas Chemical Industries, Inc.
  production of ultrahigh-molecular-weight flocculents for waste-water treatment, 242
Atmosphere
  pollution by automobile exhausts, contract awarded for development of radioisotopic technique for measurement of, 243
  pollution by various combustion processes, use of $^{12}C/^{13}C$ ratio as indicator of, 381–85
Atomchem Corp.
  purchase of hot-cell equipment for encapsulating ^{90}Sr and ^{137}Cs sources, 241
Australia
  developments in radioisotope-powered cardiac pacemaker, 361–62
  production and uses of radioisotopes, 124–25
Automation Industries, Inc.
  production of ^{192}Ir and ^{60}Co sources for industrial radiography, 241
Automobiles
  exhaust analysis for lead by X-ray fluorescence, 163–65
  exhaust pollutants, Kryptonate-based instrument for detecting, 403–6
  exhaust pollutants, nonnuclear methods for detecting, 406–8
  pollution of atmosphere, contract awarded for development of radioisotopic technique for measurement of, 243

B

Barium-131
  availability, 240
Barley
  mutants, list of radiation produced, 97–98
Beacons
  development of ^{90}Sr acoustical for ocean installations, 131–32
Beans
  mutants, list of radiation produced, 99
Beef
  radiopreservation, 80
Bell peppers
  radiopreservation, 80
Beta sources
  fabrication of strontium silicate beads for, 222
Books and booklets
  "Californium-252 Progress", 129–30
  "Fundamentals of Radiation Protection", review, 132
  Handbook of Radionuclides, review of, 132–33
  "Medical Radionuclides: Radiation Dose and Effects", review of, 349
  "Molecular Radiation Biology", review of, 349
  "Nuclear Electronics", review of, 247–48
  "Polymer–Cement Concretes and Polymer–Concretes", review of, 248–49
  "Radiation Sensitivity of Toxins and Animal Poisons", review of, 349
  "Radioactivity Calibration Standards", 463–64
  "Specifications for Radioluminous Time Measurement Instruments. Part I. Instruments Bearing Radioactive Luminous Materials", 128
Botulism
  hazard in radiation-pasteurized fish, proposed method for testing, 211–17
Bromine
  determination in Chicago air by neutron activation analysis, 460
Bromine-82
  availability, 240
  use in flow measurements, 294–95, 297
  price reduction, 242

C

Cadmium-108
  availability, 126
Cadmium-113
  availability, 126
Cadmium-114
  availability, 126
Calcium
  detection in Chicago air by neutron activation analysis, 460
  determination by X-ray-fluorescence analysis, 158–59

This index was prepared by Theodore F. Davis of AEC's Division of Technical Information Extension, Oak Ridge, Tenn.

XUM

Calcium-45
availability, 365
Californium-252
availability to educational institutions for research purposes, 129, 463
availability for purchase, 129
information center, establishment at Savannah River Plant, 366
use in exploration of ocean bottom for minerals, 247
use in nuclear probe for detecting minerals on ocean floors, 366
use in portable neutron camera for neutron radiography, 69–74
use in simple neutron radiography unit, 318
"Californium-252 Progress"
availability, 129–30
Canada
food-irradiation program, 368–69
Cancer
neutron therapy, development of neutron source from the fusion of ^{2}H and ^{3}H for, 348
Carbohydrates
structure of, study by ^{13}C nuclear magnetic resonance spectroscopy, 319
Carbon-12
price reduction, 242, 463
Carbon-13
price reduction, 242
study of carbohydrate structure by nuclear magnetic resonance spectroscopy of, 319
Carbon-14
availability, 365
labeled compounds, availability of, 241
use in comparing metabolism in man and animals, 473–74
use in dating of iron, 320
Carbon dioxide
metabolism by soil microorganisms, use of radioisotopes in study of, 103–5, 109–10
properties of, 255
use as indicator of air pollution by combustion processes, 381–85
Carbon monoxide
detection in automobile exhausts, Kryptonate-based instrument for, 403–6
Cardiac pacemaker (radioisotope powered) (see Heart)
Carnations
mutants, list of radiation produced, 100
Castor bean
mutants, list of radiation produced, 100
Cellulose acetate films
diffusion of H_2O, Na ions, and Cl ions through, radiochemical study of, 60–64
Cellulose fibers
radiation grafting, 84–94
Cement
determination in concrete, portable nuclear gage for, 146–51
Cerium
determination in Chicago air by neutron activation analysis, 460
Cerium-136
availability, 240
Cerium-138
availability, 364
Cerium-142
availability, 364
Cesium
determination in Chicago air by neutron activation analysis, 460
Cesium-131
availability, 240
Cesium-137
production of encapsulated sources of, 241
Chart of the Nuclides
availability of new version, 242
Chemicals
reaction rates, use of nuclear decay rate for measurement of, 473
synthesis, use of radiation in, 219–20
Chloride ions
diffusion through protective coatings, radiochemical study of, 60–64
Chlorine
determination in Chicago air by neutron activation analysis, 460
Chlorine-36
availability, 365
Chlorophyll
metabolism in green plants, tracer studies of, 474–75
Chromium
determination in Chicago air by neutron activation analysis, 460
determination by X-ray-fluorescence analysis, 158–59
Chromium-50
availability, 364
Chromium-51
use in flow measurements, 295–96
Chromium salt
radiation-induced reduction from hexavalent to trivalent state in hide tanning process, 222
Chrysanthemum
mutants, list of radiation produced, 100
Circuits
micro-, fabrication by neutron-transmutation doping of Si, 46–60
Citrus leaves
radiodisinfestation, 80
Clams
cryogenic radiosterilization, 81–82
Clothes
radiodisinfestation, 80
Cobalt
determination in Chicago air by neutron activation analysis, 460
Cobalt-60
evaluation of Texas A & M 40,000-Ci gamma source, 186
heat-source availability on loan, 128
loan to University of Massachusetts for food-preservation research, 129
production and delivery of millionth curie of, 246–47
production of industrial radiography sources of, 241
resistance tests for sealed teletherapy sources of, 472
standard source of, 127
use in flow measurements, 295–96
Cod fish
cryogenic radiosterilization, 81–82
maximum shelf life of irradiated and nonirradiated fillets of, 322–23
radiation effect on chemical composition of, 323–37
Computers
use in design of stacks, 460–61
Concrete
portable nuclear gage for measuring cement content in, 146–51
radiography inspection, 470
Concrete–plastic combinations
book on, review of, 248–49
development, 221
development for highway applications, 152–53
use of sewage waste and refuse as aggregate in, 243
Conferences and meetings
(see also Symposia)
"MEDEX-71", 465–66
on animal protein production from nonprotein nitrogen, 468–69
on isotopes and radiation in parasitology, 467–68
on production and uses of neutron sources, schedule for, 246
on research materials for nuclear measurements, 466
on rice breeding with radiation-induced mutations, 347
on sterilization and preservation of biological tissues by ionizing radiation,
Copper
determination by X-ray-fluorescence analysis, 158–59
Copper-64
availability, 240
Copper wire
labeling with ^{110m}Ag for subsequent identification, 315–17
Corrosion
protective coatings, radiochemical study of diffusion of ions through, 60–64
Cosmetics
radiosterilization, 131
Cotton textiles
radiation grafting, 84–94
Crab
radiopreservation, 78
Criminology
labeling of copper wire with ^{110m}Hg for subsequent identification, 315–17
Cucumbers
radiopreservation, 80

D

Dahlia
mutants, list of radiation produced, 101
Dates
radiopreservation, 80
Densimeters (alpha-particle)
for measuring the density of the Martian atmosphere, 185–86
Diseases, physiological
myocardial infarctions, prediction by hemodynamic evaluation using ^{131}I, 111–13
Douglas fir
radiopreservation, 78
Dysprosium-160
availability, 364
Dysprosium-161
availability, 126
Dysprosium-163
availability, 126
Dysprosium-164
availability, 126

E

Educational institutions
availability of ^{252}Cf for research purposes, 129
Eggplant
radiopreservation, 80
Elastomers
radioisotope methods of analysis and characterization of, 13–34
Electron accelerators
for radiation processing, design activities in the USSR of, 222

XUM

Epoxy MIL-C-22750 films
diffusion of H_2O, Na ions, and Cl ions through, radiochemical study of, 60–64
Erbium-162
availability, 364
Erbium-164
availability, 462
Erbium-166
availability, 240
Ethane, 1,2-dichloro-
radiation-initiated chlorination of, 219
Europium
determination in Chicago air by neutron activation analysis, 460
Evergreens
radiopreservation, 78
Exhibits
Life Science Radiation Laboratory, showing at Columbus, Ohio, Center of Science and Industry, 366
Your Stake in the Atom, showing at the Central Wyoming Fair and Rodeo, Casper, Wyo., 366–67

F

Fallout
production of synthetic, 408–12
Figs
radiopreservation, 79
Firestone Tire and Rubber Company of Akron, Ohio
purchase of Radiation Dynamics, Inc., 464
Fish
analysis for metals by neutron activation analysis for pullution control, 129, 460
botulism hazard in radiation-pasteurized, proposed method for testing, 211–17
cryogenic radiosterilization, 81–82
maximum shelf life of irradiated and nonirradiated cod and haddock, 321
radiation effect on chemical composition of cod and haddock, 323–37
Fish extract
radiopreservation, 78
Fish pellets
radiopreservation, 78
Flowmeters
review of conventional, 287–91
review of radioisotopic, 291–307
Fluorescent dyes
use in estuary pollution-control studies, comparison with radioisotopes for, 385–88
Foods
(see also specific foods, e.g., Fish, Potatoes, etc.)
botulism hazard in radiation-pasteurized fish, proposed method for testing of, 211–17
irradiation program in Canada, 368–69
microbiological specifications and testing methods for irradiated, 470–71
radiation inhibition of potato sprouting, 187–206, 210
radiation processing, increase of research funds for, 243
radiopasteurization, 321–38
radiopreservation, loan of ^{60}Co for research in, 129
radiopreservation, review of the Portable Cesium Irradiator demonstrations, 75–81
training manual on irradiation technology and techniques, 471–72
use in Israel and the Netherlands, 465
wholesomeness study, 464
Forensic science
labeling of copper wire with ^{110m}Ag for subsequent identification, 315–17
France
developments in radioisotope-powered cardiac pacemaker, 360–61
facility for fabrication of sealed radiation sources, 121–22
Frostbite
evaluation with ^{131}I-RISA, 348
Fruits
radiopreservation, 80

G

Gadolinium-154
availability, 240, 364
Gages (nuclear)
for measuring cement content in concrete, design of portable, 146–51
for measuring density of Martian atmosphere, 185–86
for measuring density of soils, 165
for measuring moisture content, 469
for measuring SO_2 in stack gases, 397–402
for measuring solids content in pulps in closed pipe systems, 473
for pollution control, 395–97
^{147}Pm X-ray for determining particle-size distributions, 317–18
safety testing and standards, 116–17
for width control of layflat plastic tubing for packaging, 69
X-ray fluorescence, 153–59, 465
Gamma radiography
of concrete, 470
production of ^{192}Ir and ^{60}Co sources for, 241
using photonuclear reactions, 320
Gamma sources
evaluation of Texas A & M 40,000-Ci ^{60}Co monochromatic, 187
Germanium
neutron-transmutation doping, 37–46
Gold
determination by X-ray-fluorescence analysis, 158–59
Gold-198
availability, 240
use in flow measurements, 295–96
Goldblith, Samuel A.
recipient of Nicholas Appert Award, 249
Grain
analysis for amino acids by X-ray photoelectron spectroscopy, 369
radiodisinfestation, 79
Grand fir
radiopreservation, 78
Grapefruit
radiopreservation, 80
Guinn, Vincent P.
appointment to the Advisory Committee on Isotopes and Radiation Development, 368

H

Haddock
maximum shelf life of irradiated and nonirradiated fillets of, 323
radiation effect on chemical composition of, 323–37
Harwell Isotope Bureau
establishment of, 464
Hastelloy alloy X
compatibility with various ^{90}Sr fuel forms, 284
Haynes alloy No. 25
compatibility with various ^{90}Sr fuel forms, 284
Heart
infarctions of human, use of ^{131}I for prediction of, 111–13
radioisotope-powered pacemaker, description of French, 102
radioisotope-powered pacemaker, developments in Australia, France, and the United Kingdom, 360–62
radioisotope powered-pacemaker, development in Japan, 465
radioisotope-powered pacemaker, performance of first implant, 464
radioisotope-powered pacemaker, progress in development, 245
Highways
deicing, development of radioisotope heat sources for, 131
development of, nuclear applications in, 146–65
Holly
radiopreservation, 77–78
Hormones
availability of ^{125}I and ^{131}I labeled, 241
Hormones (steroid)
labeling with radioisotopes for quantitative determination of, 472
Hughes Aircraft Corporation
purchase of minority interest in Accelerators, Inc., 241
Hydrocarbons
detection in automobile exhausts, Kryptonate-based instrument for, 403–6
Hydrogen
location in metals, new neutron radiograph technique for, 131
Hydrogen chloride
radiation synthesis of, 219
Hydrology
use of radioisotopes in, symposium on, 469–70

I

Imperial Chemical Industries
purchase of Triga Reactor for production of short-half-life radioisotopes, 247
Indexes
permuted title to "Isotopes and Radiation Technology", 249
Indium-114*m*
availability, 240
Industrial Nuclear Co., Inc.
production of ^{125}I- and ^{131}I-labeled proteins and hormones, 241
Information centers
on ^{252}Cf, establishment of, 366
International Nutronics, Inc.
radiosterilization activities, 127
Iodine-125
labeled proteins and hormones, availability of, 241
Iodine-131
labeled proteins and hormones, availability of, 241
use in evaluation of frostbite damage, 348
use in flow measurements, 295
use in prediction of myocardial infarctions, 111–13
Iodine-132
use in flow measurements, 295

XUM

Iridium-192
production of industrial radiography sources of, 241
Iron
^{14}C dating of, 320
determination in Chicago air by neutron activation analysis, 460
determination by X-ray-fluorescence analysis, 158–59
Irradiation facilities
availability of portable with ^{90}Sr–^{90}Y source, 461
^{60}Co, expansion of industrial, 129
Gammators, availability of, 127–28
Radtank, availability from RTI, 128
review of the Portable Cesium Irradiator demonstrations, 75–81
University of Missouri, description and applications of, 137–45
Irradiation sources
(see Beta sources, Gamma sources, and Neutron sources)
Isotope Techniques Laboratory of Stockholm
tenth year of operation, 363
Isotopes Development Division
contractor reports, list of, 371
contracts awarded, 243, 368
research and development projects for 1970, availability of progress report on, 367
research and development projects for 1969, availability of progress report on, 244
Isotopes Information Center
chemical patent search service, 126
permuted title index, 249

K

Kevex X-ray-fluorescence spectrometer
description of, 65
Krypton
isotopic composition of fission produced, 256
large-scale utilization of fission produced, 253–60
properties of, 255
recovery from air, 255–57
recovery from irradiated fuels, 256–60
Krypton-85
use in detecting pollutants in automobile exhausts, 403–6
use in flow measurements, 295–96
use in self-luminescent highway signs, 161–63

L

Lakes
evaporation mechanism in, tritiated-water-vapor method for studying, 389–91
Lanthanum
determination in Chicago air by neutron activation analysis, 460
Larson, Clarence E.
speech at the Symposium on Low Energy X- and Gamma-Ray Sources and Applications, 244
Lead
determination in automobile exhaust by X-ray-fluorescence analysis, 163–65
determination by X-ray-fluorescence analysis, 158–59
Lemon seeds
radiodisinfestation, 80
Lemons
radiopreservation, 80
Lespedeza
mutants, list of radiation produced, 100
Lightning rods
design of radioactive air ionization, 178–80
Lockheed-Georgia
closing of nuclear laboratory at Davisonville, Ga., 365
Lungs
function, ^{133}Xe analyzer for, 226–35

M

Manganese
determination in Chicago air by neutron activation analysis, 460
determination by X-ray-fluorescence analysis, 158–59
Manowitz, Bernard
appointment to the Advisory Committee on Isotopes and Radiation Development, 368
Mars
atmospheric density, alpha-particle densimeter for measurement of, 185–86
Massachusetts, University of
recipient of ^{60}Co for food-preservation research, 129
Medi-Physics, Inc.
establishment, 241
Medical products
radiosterilization, 465
Medicine
dual radioisotopic method for diagnosis of pulmonary embolism, 346–47
frostbite evaluation with ^{131}I-RISA, 348
^{133}Xe pulmonary-function analyzer, 226–35
use of radioisotopes in, survey of, 236–39
Mercury
determination in Chicago air by neutron activation analysis, 460
determination in fish by neutron activation analysis, 460
determination of trace amounts by neutron activation analysis, 461
Mercury-197
availability, 240
Mercury-198
availability, 240
Mercury-203
standard source of, 127
Metabolism
comparison in man and animals, use of ^{14}C in, 473–74
of CO_2 by soil microorganisms, radiometric study of, 103–5, 109–10
Metallography
uses of ^{57}Fe Mossbauer effect in, 166–74
Metals
new neutron radiograph technique for locating low concentrations of H_2 in, 131
radioactive-tracer-diffusion data, compilation on, 471
Microcircuits
fabrication by neutron-transmutation doping of semiconductors, 46–60
Microorganisms
metabolic reactions in soils, use of radioisotopes in study of, 103–10
Minerals
detection on ocean floors, use of ^{252}Cf in, 247, 366
Missouri, University of
Research Reactor Facility, description and applications of, 137–45
Moisture
neutron gages for measuring, guidebook on theory and practices, 469
Molybdenum
compatibility with various ^{90}Sr fuel forms, 284
determination by X-ray-fluorescence analysis, 158–59
Molybdenum-99
availability, 240, 462
availability of 99.99% pure, 365
Moon
development of portable neutron activation analysis device for use on, 126
Mossbauer effect
potential industrial applications of ^{57}Fe, 166–74
Mud–plastic combinations
development, 221
Mushrooms
radiopreservation, 78–79
Mutations
breeding in plants, manual on, 466
radiation induced, list of benificial, 96–101
Mylar films
diffusion of H_2O, Na ions, and Cl ions through, radiochemical study of, 60–64

N

N-155 alloy
compatibility with various ^{90}Sr fuel forms, 284
National Bureau of Standards
availability of standard radiation sources from, 127
Navigational aids
^{90}Sr-powered acoustical beacons for ocean installations, 131–32
Neodymium-142
future availability, 240, 364
Neodymium-143
future availability, 240, 364
Neodymium-144
future availability, 240
Neodymium-145
future availability, 240, 364
Neodymium-148
future availability, 240, 364
Neodymium-150
future availability, 240, 364
Neon
properties of, 255
Neoprene 1672A films
diffusion of H_2O, Na ions, and Cl ions through, radiochemical study of, 60–64
Neutron activation
use in recovery of faded photographs, 309–14
Neutron activation analysis
development of portable instrument for analysis of moon and other solar bodies, 126
of fish for metals for pollution control, 129, 460
in oceanography, review of symposium on, 175–78
of oil slicks for identification, 377–79
use in detection of various elements in Chicago air, 460

XUM

Neutron moisture gages
guidebook on theory and practice, 469
Neutron Products, Inc.
sale of millionth curie of ^{60}Co, 246–47
Neutron radiography
design and applications of portable neutron camera for, 69–74
design of simple unit using ^{252}Cf, 318
new technique for locating low concentrations of H_2 in metals, 131
Neutron sources
availability of Ra–Be, Sb–Be, Pu–Be, and Th–Be, 241–42
development from fusion of ^{2}H and ^{3}H for cancer therapy, 348
production and uses of, schedule of meeting on, 246
Neutrons
interactions with materials, secondary gamma production by, 132
Nickel
determination by X-ray-fluorescence analysis, 158–59
Nickel-58
availability, 240
Nickel-61
availability, 240
Nickel-62
availability, 240
Nickel-63
availability, 365
standard source of, 127
Nickel-64
availability, 126
Niobium
compatibility with various ^{90}Sr fuel forms, 284
determination by X-ray-fluorescence analysis, 158–59
Ni-o-nel
compatibility with various ^{90}Sr fuel forms, 284
Nitrocellulose acrylic lacquer films
diffusion of H_2O, Na ions, and Cl ions through, radiochemical study of, 60–64
Nitrogen
metabolism by soil microorganisms, use of radioisotopes in study of, 103–5, 109–10
Nitrogen-15
labeled compounds, availability of, 241
Nitrogen oxides
detection in automobile exhausts, Kryptonate-based instrument for, 403–6
Nuclear gages
(see Gages (radiometric))
Nuclear magnetic resonance spectroscopy
method of studying carbohydrate structure using ^{13}C, 319
Nuclear probes
development for detecting minerals on ocean floors, 366
Nucleic acid
metabolism by soil microorganisms, use of radioisotopes in study of, 103–5, 109–10
Nylon 8200 films
diffusion of H_2O, Na ions, and Cl ions through, radiochemical study of, 60–64

O

Oak Ridge National Laboratory
availability of stable isotopes from, 126, 240, 364, 462
electromagnetic separations program, 240–41, 364–65, 462–63
Oats
mutants, list of radiation produced, 98
Oceanography
use of neutron activation analysis in, review of symposium on, 175–78
Oil slicks
identification of, use of activation-analysis and sulfur-isotope-ratio techniques for, 379–81
Orange juice
radiopreservation, 80
Oranges
radiopreservation, 75, 80
Oxygen-16
price reduction, 242, 463
Oxygen-17
availability, 364, 462
labeled compounds, availability of, 241
Oxygen-18
availability, 240, 364
labeled compounds, availability of, 241
Ozone
high-energy radiation synthesis of, 420–27

P

Packaging material
layflat plastic tubing, radioisotopic gage for width control of, 69
Paintings
forgeries, detection by X-ray-fluorescence spectroscopy, 65–68
Panametrics
development of radioisotopic technique to determine air pollution from automobile exhausts, 243
Paper
wet strength improvement by radiation grafting of, 222
Paraffins
radiation-induced sulfoxidation of, 220
Parasitology
use of isotopes and radiation in, meeting on, 467–68
Particles
particle-size distributions, radioisotopic X-ray gage for determining, 317–18
Patents (chemical)
search service by Isotopes Information Center, 126
Peaches
mutants, list of radiation produced, 100
radiopreservation, 79–80
Peanuts
mutants, list of radiation produced, 99
Peas
mutants, list of radiation produced, 99
Pesticides
nuclear techniques for studying residue problems, 459
radioisotopes in research on, 459
regional variation in percutaneous penetration in man, 459
Phosphorus-32
availability, 365
Photographs (faded)
recovery by neutron activation, 309–14
Plastic–concrete combinations
book on, review of, 248–49
development, 221
development for highway applications, 152–53
use of sewage waste and refuse as aggregate in, 243
Plastic–mud combinations
development, 221
Plastic–red mud combinations
development, 221
Plastic tubing
layflat, radioisotopic gage for width control of, 69
Plastic–wood combination
development, 221
production facilities at RTI, expansion of, 128
use in restoration of French parquet floor, 472–73
Plutonium-238
price reduction, 243
production in Connecticut Yankee Reactor, 463
standard source of, 127
use in cardiac pacemaker, 360–61
use as heat source for integrated waste management – water recovery system on space vehicles, 130
use as power supply in the Transit navigation satellite, 130
Pollution
(see also specific pollutants and specific polluted environments, e.g., Sulfur dioxide, Surface waters, etc.)
control, radioisotope gages for, 377–78
measurement and control, symposium on nuclear techniques for, 377–78
Polyethylene films
diffusion of H_2O, Na ions, and Cl ions through, radiochemical study of, 60–64
Polymerization
use of radiation in, 220–21
Portable Cesium Irradiator
demonstrations, review of, 75–81
Potassium
determination by X-ray-fluorescence analysis, 158–59
Potatoes
radiopreservation, 75, 79–80
sprouting, inhibition by radiation, 187–206, 210
Poultry wastes
radiation processing of, 223–24
Powders
particle-size distributions, radioisotopic X-ray gage for determining, 317–18
Precipitation
separation of ^{90}Sr from Purex wastes by, 265–67
Promethium-147
use in self-luminous dials, specifications for, 128
use in wristwatch, 464
Protective coatings
diffusion of ions through, radiochemical study of, 60–64
electron-beam curing of, 220–21
water permeability, measurement by radioisotope tracer technique, 181–84
Proteins
analysis for amino acids by X-ray photoelectron spectroscopy, 369
availability of ^{125}I and ^{131}I labeled, 241
production by ruminants from non-protein nitrogen, report on, 468–69
Prunes
radiopreservation, 80
Pulmonary embolism
diagnosis, dual radioisotopic method for, 346–47

R

Radiation detectors
symposium on new developments in physical and biological, 127
Radiation Dynamics, Inc.
purchase by Firestone Tire and Rubber Company of Akron, Ohio, 464

XUM

Radiation processing
chemical synthesis, 219–20
dosimetry studies, 221–22
engineering studies, 222
polymerization and modification of polymers, 220–21
of poultry wastes, 223–24
review of symposium on, 219–22
Radiation protection
book, review of, 132
Radiation sources
(see also Beta sources, Gamma sources, and Neutron sources)
book, "Radioactivity Calibration Standards", 463–64
for calibration of radiation detectors, availability of NBS standard, 127
French facility for fabrication of sealed, 121–22
Radiation Technology, Inc.
irradiation services and products, 128–29
Radioactive materials
design and evaluation of shipping containers for, 350–59
Radioactive wastes
Purex 1WW, composition of, 265
Purex 1WW, separation of ^{90}Sr from, 264–68
Radiography
(see Gamma radiography and Neutron radiography)
Radiography equipment
inspection and maintenance of, amendment of regulations on, 367–68
safety testing and standards, 114–16
Radioisotope heaters
development for aerospace applications, contract awarded for, 243
use in highway-deicing systems, 131
Radioisotope-powered electric generators
development for use on the Navy's advanced Transit navigation satellite, 130
development for use in producing optical and acoustic signals for offshore oil rigs, 465
IAEA safety design manual for, 249
LCG 25A, status of, 262
LCG 25B, status of, 262
LCG 25CI, status of, 263
Marguerite 20, 133
MW-3000A, status of, 262
NUMEC 100 MA(2) milli TRACS, status of, 262
NUMEC TRACS-25A, status of, 262
orders for ^{90}Sr, 245
PI-1003, status of, 263
PI-1004, status of, 263
PI-1005, status of, 263
PI-1006, status of, 263
RIPPLE III, status of, 263
RIPPLE IV, status of, 263
RIPPLE IX, status of, 263
Sentinel 25D, status of, 263
Sentinel 25E, status of, 263
Sentry, status of, 262
SNAP-3A, performance, 366
SNAP-7A, examination of $^{90}SrTiO_3$ fuel from, 280–83
SNAP 7A, status of, 262
SNAP 7B, status of, 262
SNAP 7C, status of, 262
SNAP 7D, status of, 262
SNAP 7E, status of, 262
SNAP 7F, status of, 262
SNAP-21, status of, 262
SNAP-23A, status of, 262
UI-1001, status of, 262
URIPS(PI-1001A), status of, 262
URIPS(PI-1002), status of, 262
use of ^{90}Sr as fuel in, review of, 260–84
2-kW(e) thermoelectric, ORNL development program for, 345
3-kW(e) steam Rankine cycle, ORNL development program for, 344–45
5-kW(e) organic Rankine cycle, ORNL development program for, 341–44
Radioisotope tracer techniques
labeling of copper wire with ^{110m}Ag for subsequent identification, 315–17
use in analysis and characterization of elastomers, 13–34
use in chemical rate measurement, 473
use in flow measurements, 291–302
use in measuring water permeability of protective coatings, 181–84
use in metabolism studies, 103–10, 473–75
use in sand-movement studies, 392–94, 412–14
use in stream pollution studies, 472
Radioisotopes
25th anniversary of the first shipment of, comments by Glenn T. Seaborg on, 373–76
availability of inexpensive shipping container for, 365
packaging guide, 244
production by spallation, data compilation on, 1–12
production from fission products, review of, 474
production techniques, seminar on, 243
production and uses in Australia, 124–25
separation program at ORNL, 240, 364, 464
use in agriculture, publication on, 466–67
use in animal science and veterinary medicine, training course on, 365
use in flow measurement, review of, 287–307
use in industry, study by Latin-American and Asian scientists, 369–70
use in Japan, increase in, 466
Radioisotopic gages
(see Gages (nuclear))
Radioluminous materials
use on instruments, specifications for, 128
Radiopharmaceuticals
specifications, brochure on, 242
Radiopreservation
of biological tissues, conference on, 469
of fishery products, 321–38
of foods, wholesomeness study of, 464
of potatoes, 187–206, 210
review of Portable Cesium Irradiator demonstrations, 75–81
Radiosterilization
of biological tissues, conference on, 469
of cosmetics, 128, 131, 462–63, 465
of enzymes, 128
of enzymes for detergents, 127
of medical supplies, 128, 462–63
pilot plant for sewage processing by, 127
plants, European purchase from AECL, 131
of tubes of distilled water in blood-test kit, 127
Radiotherapy
development of neutron source for treatment of cancer, 348
high-energy centers, directory of, 365
resistance tests for sealed ^{60}Co sources for, 472
Radium-226
use in self-luminous dials, specifications for, 128
Radwood
production facilities at RTI, expansion of, 128
Rainbow trout
radiopreservation, 78–79
Raisins
radiopreservation, 79
Reactors
IAEA comprehensive list of, 468
Reactors (Connecticut Yankee)
production of ^{238}Pu, 463
Reactors (University of Missouri Research)
description and applications of, 137–45
Red mud–plastic combinations
development, 221
Red snapper
radiopreservation, 78–79
Respiratory system
(see Lungs)
Rhenium-185
availability, 364
Rhenium-187
availability, 364
Ribonuclease acid
decomposition of, radiometric study of, 473
Rice
fertilization studies, review of IAEA report on uses of radioisotopes in, 347
mutants, list of radiation produced, 98
radiation-induced mutations in, report of meeting on, 347
radiopreservation, 80
Rivers
(see Surface waters)
Roses
mutants, list of radiation produced, 100
Rubber
radioisotope methods of analysis and characterization of, 13–34

S

Salmon
radiopreservation, 78–79
Sand
detection system for tracing radioisotope labeled, 412–14
labeling with radioisotopes for studying movement of, 392–94
labeling with radioisotopes for synthetic-fallout studies, 408–12
Seaborg, Glenn T.
comments on 25th anniversary of the first radioisotope shipment, 373–76
Selenium
determination in Chicago air by neutron activation analysis, 460
Selenium-78
availability, 126
Selenium-82
availability, 126
Self-luminescent materials
use in highway signs, 161–63
Semiconductor devices
fabrication by neutron-transmutation doping of Ge and Si, 37–46
Seminars
on radioisotope-production techniques, 243
Sewage waste
radiation processing of, 222, 429–33, 435–44, 449–50, 452–53

XUM

use as aggregate in concrete–plastic products, 243
Shielded containers
for small volumes of radioactive solutions, 242
Shipping
of radioactive materials, design and evaluation of containers for, 350–59
Shrimp
radiopreservation, 79–80
Shrimp (brown)
radiation effect on chemical composition of, 323–37
Shrimp (pink)
radiation effect on chemical composition of, 323–37
Signs
use of self-luminescent materials in, 161–63
Silicon
neutron-transmutation doping, 37–60
Silver
determination by X-ray-fluorescence analysis, 158–59
Silver-110*m*
use in labeling copper wire for subsequent identification, 315–17
Sitka spruce
radiopreservation, 78
Sodium
determination in Chicago air by neutron activation analysis, 460
Sodium-22
standard source of, 127
Sodium-24
availability, 240
use in flow measurements, 294–95
Sodium ions
diffusion through protective coatings, radiochemical study of, 60–64
Soils
density measurement by gamma-ray backscattering, reduction of errors in, 165
metabolic studies of microorganisms in, use of radioisotopes for, 103–10
Solvent extraction
separation of ^{90}Sr from Purex wastes by, 267–69
Soybeans
mutants, list of radiation produced, 98
Space missions
integrated waste management – water recovery system for, development of radioisotope powered, 130
Spallation
production of radioisotopes by, data compilation on, 1–12
Spectroscopy
(see X-ray-fluorescence spectroscopy)
Spring rape
mutants, list of radiation produced, 99
Stable isotopes
availability from ORNL, 128, 240, 364
Stack gases
analysis for SO_2, nonnuclear methods for, 402–3
analysis for SO_2, radioisotopic instrument for, 397–402
Stacks
design of, computer program for, 460–61
Strawberries
radiopreservation, 75
Streams
(see Surface waters)
Streptocarpus
mutants, list of radiation produced, 101
Strontium-90
chemical and physical properties of, 283
compatibility with various container materials, 284
fabrication for beta sources, 222
maximum permissible body burdens for, 283–84
maximum permissible concentrations in air and water for occupational exposure for, 283–84
production of encapsulated sources of, 241
separation from Purex wastes, 264–68
use in acoustical beacons for ocean installations, 131–32
use as fuel in thermoelectric generators, review of, 260–84
Strontium disilicide ($^{90}SrSi_2$)
compatibility with various container materials, 284
Strontium fluoride ($^{90}SrF_2$)
chemical and physical properties of, 283
compatibility with various container materials, 284
Strontium oxide (^{90}SrO)
chemical and physical properties of, 283
compatibility with various container materials, 284
fabrication into heat sources, 272–80
preparation of, 271
Strontium oxide systems (^{90}SrO–BeO)
compatibility with various container materials, 284
Strontium titanate ($^{90}SrTiO_3$)
chemical and physical properties of, 283
compatibility with various container materials, 284
fabrication into heat sources, 272–80
preparation of, 268, 270–71
from SNAP-7A, examination of, 280–83
Strontium titanate ($^{90}Sr_2TiO_4$)
chemical and physical properties of, 283
Sulfur
determination by X-ray-fluorescence analysis, 158–59
metabolism by soil microorganisms, use of radioisotopes in study of, 103–5, 109–10
Sulfur-33
availability, 126, 240
Sulfur-35
availability, 365
Sulfur dioxide
determination in stack gases, nonnuclear methods for, 402–3
determination in stack gases, radioisotopic instrument for, 397–402
Suntac Nuclear Corporation
establishment, 241
Surface waters
pollution, tracer studies of, 472
pollution by paper-mill activities, contract awarded for development of radioisotopic techniques for measurement of, 243
pollution-control studies, comparison of radioisotopes and fluorescent dyes in, 385–88
Symposia
(see also Conferences and meetings)
on biophysical aspects of radiation quality, schedule for, 245
on inhibition of potato sprouting by radiation, 210
on isotope hydrology, 469–70
"Medical Radionuclides: Radiation Dose and Effects", 136
on neutron activation analysis in oceanography, review of, 175–78
on nuclear techniques for measurement and control of environment pollution, 377–78
on new developments in physical and biological radiation detectors, 127
packaging and transportation of radioactive materials, schedule for, 246
on use of radiation and radioisotopes for genetic improvement of industrial microorganisms, schedule for, 245–46
on utilization of large radiation sources and accelerators in industrial processing, review of, 219–22

T

Tantalum
compatibility with various ^{90}Sr fuel forms, 284
Tellurium-122
availability, 462
Tellurium-125
future availability, 240, 364, 462
Tellurium-126
future availability, 240, 364, 462
Texas A & M University
development of portable neutron activation analysis device for use on the moon and other solar bodies, 126
Textiles
radiation processing of, 220
Time measurement
(see Wristwatch)
Tin
determination by X-ray-fluorescence analysis, 158–59
Tin-112
availability, 240, 462
future availability, 240, 364, 462
Tin-114
future availability, 240, 364, 462
Tin-115
future availability, 240, 364, 462
Tin-117
future availability, 240, 364, 462
Tin-118
future availability, 240, 364, 462
Tin-119
future availability, 240, 364, 462
Tin-120
future availability, 240, 364
Tin-122
future availability, 240, 364, 462
Tin-124
future availability, 240, 364, 462
Titanium
determination by X-ray-fluorescence analysis, 158–59
Tobacco
mutants, list of radiation produced, 100
Tomatoes
radiopreservation, 75, 80
Tooth enamel
density measurement of, development of radioisotopic radiation-backscatter gage for, 243
Tracer techniques
(see Radioisotope tracer techniques)
Training courses
on advanced techniques involving radioisotopes and radiation in animal science and veterinary medicine, 365

XUM

Transportation
of radioactive materials, design and evaluation of containers for, 350–59
Tritium
labeled compounds, availability of, 241
use in flow measurements, 295–96
use in self-luminous dials, specifications for, 128
Tungsten
compatibility with various ^{90}Sr fuel forms, 284
determination by X-ray-fluorescence analysis, 158–59
TZM alloy
compatibility with various ^{90}Sr fuel forms, 284

U

Union of Soviet Socialist Republics
development of electron accelerators, 222
United Kingdom
developments in radioisotope-powered cardiac pacemaker, 360–61
United Kingdom Atomic Energy Authority
establishment of Harwell Isotope Bureau, 404
recipient of the Queen's Award to Industry, 369
Uranium
determination by X-ray-fluorescence analysis, 158–59
Universal Radioisotopes, Inc.
production of Ra–Be, Sb–Be, Pu–Be, and Th–Be neutron sources, 241–42

V

Vanadium
determination by X-ray-fluorescence analysis, 158–59
Veterinary medicine
use of radioisotopes in, training course on, 365
Vulcanization
use of radiation in, 221

W

Washington State University, Pullman
study of radioisotopic techniques to determine stream pollution from paper-mill activities, 243
Water
diffusion through protective coatings, measurement by radioisotope tracer technique of, 181–84
diffusion through protective coatings, radiochemical study of, 60–64
recovery system for space missions, development of radioisotope powered, 130
Water (waste)
availability of ultrahigh-molecular-weight flocculents for treatment of, 242
high-energy radiation synthesis of ozone for treatment of, 420–27
radiation processing of, 415–20, 429–33, 435–38, 444–49, 451, 453–57
Western hemlock
radiopreservation, 78
Wheat
mutants, list of radiation produced, 97
White mustard
mutants, list of radiation produced, 99
Wiggin, Edwin A.
appointment to the Advisory Committee on Isotopes and Radiation Development, 368
Wood–plastic combination
development, 221
production facilities at RTI, expansion of, 128
use in restoration of French parquet floor, 472–73
Wristwatches
development of ^{147}Pm powered, 464

X

X-ray-fluorescence analyzers
availability of, 465
description and uses of commercially available portable radioisotope, 153–59
for measuring lead in automobile exhaust, 163–65
X-ray-fluorescence spectroscopy
detection of art forgeries by, 65–68
report on, 474
X-ray photoelectron spectroscopy
use in grain analysis for amino acids, 365
Xenon
isotopic composition of fission produced, 256
large-scale utilization of fission produced, 253–60
properties of, 255
recovery from air, 255–57
recovery from irradiated fuels, 256–60
Xenon-128
production from ^{127}I, 244
Xenon-133
availability for injections in disposable syringes, 461
use in flow measurements, 295–97
use in pulmonary-function analysis, 226–35

Y

Ytterbium-169
availability, 240
Yttrium-88
standard source of, 127

Z

Zinc
determination in Chicago air by neutron activation analysis, 460
determination by X-ray-fluorescence analysis, 158–59
Zoller Dental Clinic
development of radioisotopic radiation-backscatter gage for tooth-enamel-density measurements, 243

XUM

AEC Symposium Series

PRECIPITATION SCAVENGING (1970)

Coordinators: R. J. Engelmann and W. G. N. Slinn

This volume contains the proceedings of the 1970 Precipitation Scavenging Meeting, Battelle–Northwest, Richland, Washington, June 2–4, 1970. Thirty-eight papers assess the state-of-the-art and provide a wide perspective of research and projects, including experimental techniques, experimental data, microphysics in scavenging, and models for predicting scavenging. Available as CONF-700601 for $6.00 a copy from the National Technical Information Service, Springfield, Virginia 22151. (512 pages, 194 illus., 6 by 9 in., Library of Congress Catalog Card No. 70-609397)

CONTENTS

Scavenging in Perspective, *J. J. Fuquay*

Priorities in Scavenging Research, *R. J. Engelmann*

Radar Observations of Convective Storm Circulation and Its Relation to Precipitation Scavenging, *R. M. Lhermitte*

Scavenging of Tracer in Severe Storms, *A. N. Dingle*

Interactions of Cloud Condensation Nuclei and Ice Nuclei with Cloud and Precipitation Elements: A Review, *L. F. Radke, W. D. Scott, and C. E. Robertson*

Entry of Freezing Nuclei into Precipitation, *G. Vali*

In-Cloud Scavenging Analysis from Cosmogenic Radionuclide Measurements, *R. W. Perkins, C. W. Thomas, J. A. Young, and B. C. Scott*

Stable Elements of the Atmosphere as Tracers of Precipitation Scavenging, *L. A. Rancitelli, R. W. Perkins, T. M. Tanner, and C. W. Thomas*

Concentration Variation of Some Trace Metals in Precipitation from Great Plains Thunderstorms, *D. H. Steffe*

Scavenging by Snow and Ice Crystals, *S. K. Sood and M. R. Jackson*

Scavenging of Soluble Dye Particles by Rain, *M. T. Dana*

Collection Efficiencies of Raindrops for Submicron Particulates, *J. R. Adam and R. G. Semonin*

Collection Efficiency of Water Droplets in AgCl Aerosol, *M. Kerker, E. Matijević, V. Hampl, and D. Cooke*

Collection Efficiency in Washout by Rain, *T. G. Owe Berg*

Scavenging of Aerosol Particles by Sprays, *J. D. McCormack and R. K. Hilliard*

Continuous Charged Cloud-Particle Sampler, *C. P. R. Saunders, M. H. Smith, and J. Latham*

Pulsed-Laser Holography for Analysis of Particle Size and Distribution, *F. R. Reich and B. P. Hildebrand*

Thermal-Diffusion Chambers as Cloud-Nuclei Counters, *V. K. Saxena and J. L. Kassner, Jr.*

Automatic Sequential Rain Sampler for Scavenging Studies, *D. F. Gatz, R. F. Selman, R. K. Langs, and R. B. Holtzman*

Effect of Solubilities of Gases on Their Scavenging by Raindrops, *A. K. Postma*

Laboratory Investigations on Washout of Trace Gases, *S. Beilke*

Washout of Gaseous Iodine Species, *L. F. Coleman and A. K. Postma*

European Interest in Acidic Precipitation, *H. Reiquam*

Washout of SO_2 from the Plume of a Coal-Fired Power Plant, *J. M. Hales, J. M. Thorp, and M. A. Wolf*

Scavenging of SO_2 by Convective Storms, *P. W. Summers*

Mechanisms of Iodine Injection from the Sea Surface, *C. S. Martens and R. C. Harriss*

Model for Variations with Particle Size of Halogen-Ion Ratios in Marine Aerosols, *J. A. Robbins*

Theory of Diffusive and Impactive Scavenging, *G. M. Hidy*

Attachment of Trace Substances on Atmospheric Aerosols, *J. R. Brock*

Recent Calculations of Collision Efficiencies, *L. M. Hocking*

Processes Influencing Evolution of Droplet or Aerosol Spectrum, *R. L. Drake*

Particle-Free Space in Stefan Flow, *O. A. Vittori and V. Prodi*

Phoretic Processes in Scavenging, *W. G. N. Slinn and J. M. Hales*

Precipitation Scavenging in a Large-Cloud Diffusion Code, *K. R. Peterson and T. V. Crawford*

Scavenging of Aerosols by Rain: A Numerical Study, *R. N. Kortzeborn and F. F. Abraham*

Tropospheric Scavenging of ^{90}Sr and ^{3}H, *P. W. Krey and B. Krajewski*

Use of Natural Radioactivities To Estimate Large-Scale Precipitation Scavenging, *L. Machta, R. J. List, M. E. Smith, Jr., and H. Oeschger*

Scavenging Prediction Using Ratios of Concentrations in Air and Precipitation, *R. J. Engelmann*

Closing Remarks, *R. J. Engelmann*

XUM

NUCLEAR SCIENCE ABSTRACTS

The U. S. Atomic Energy Commission, Division of Technical Information, publishes *Nuclear Science Abstracts (NSA)*, a semimonthly journal containing abstracts of the literature of nuclear science and engineering.

NSA covers (1) research reports of the U. S. Atomic Energy Commission and its contractors; (2) research reports of government agencies, universities, and industrial research organizations on a worldwide basis; and (3) translations, patents, books, and articles appearing in technical and scientific journals.

Complete indexes covering subject, author, source, and report number are included in each issue. These indexes are cumulated and sold separately.

Availability

SALE *NSA* is available on subscription from the Superintendent of Documents, U. S. Government Printing Office, Washington, D. C. 20402, at $42.00 per year for the semimonthly abstract issues and $38.00 per year for the cumulated-index issues. Subscriptions are postpaid within the United States, Canada, Mexico, and all Central and South American countries, except Argentina, Brazil, Guyana, French Guiana, Surinam, and British Honduras. Subscribers in these Central and South American countries, and in all other countries throughout the world, should remit $52.50 per year for subscriptions to semimonthly abstract issues and $47.50 per year for the cumulated-index issues. The single-copy price for the abstract issues is $1.75 postpaid, with this exception: Add one-fourth of $1.75 for mailing to the countries to which the $52.50 subscription rate applies.

EXCHANGE *NSA* is also available on an exchange basis to universities, research institutions, industrial firms, and publishers of scientific information. Inquiries should be directed to the Division of Technical Information Extension, U. S. Atomic Energy Commission, P. O. Box 62, Oak Ridge, Tennessee 37830.

XUM

Subscription Information

Technical Progress Reviews may be purchased from Superintendent of Documents, U. S. Government Printing Office, Washington, D. C. 20402. *Nuclear Safety* at $3.50 per year (six issues) for each subscription or $0.60 per issue; *Reactor Technology* at $3.00 per year (four issues) for each subscription or $0.75 per issue; *Isotopes and Radiation Technology* at $2.50 per year (four issues) or $0.70 per issue.

Postage and Remittance

Postpaid within the United States, Canada, Mexico, and all Central and South American countries except Argentina, Brazil, Guyana, French Guiana, Surinam, and British Honduras. For these Central and South American countries and all other countries: add, for each annual subscription, $1.00 for *Nuclear Safety* and $0.75 for each of the other journals; for single issues, add one-fourth of the single-issue price. Payment should be by check, money order, or document coupons, and MUST accompany order. Remittances from foreign countries should be made by international money order or draft on an American bank payable to the Superintendent of Documents or by UNESCO book coupons.

XUM

XUM